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Photo-assisted rechargeable supercapacitors and applications

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Photo-assisted rechargeable supercapacitors (PSCs) are a new category of hybrid energy-storage devices that combine solar energy harvesting and electrochemical charge storage within a single device. The review presents the latest developments in PSCs, including their operation principles, material design strategies, architectures, and performance characteristics. Specifically, the emphasis is on asymmetric and integrated architectures that use photoresponsive electrodes, in which light-generated charge carriers enhance redox reactions and ion transport. The advances in photoactive materials, including transition metal oxides and sulfides, TiO_2 nanotubes and hybrid heterostructures, are discussed in connection with the enhancement of specific capacitance, energy density, and photocharging efficiency. Moreover, the electrochemical performance in experimental studies is consistently improved under illumination compared with that under dark conditions, with energy densities in optimized systems reported to be as high as ~60.9 Wh kg^{-1} and improved coulombic efficiency. This review focuses on the importance of binder-free nanostructured electrodes, interface engineering, electrolyte optimization, and band-alignment control to enhance cycling performance and long-term stability. Theoretical and *in situ* studies have also been discussed recently to explain the process of light-initiated charge transfer. Despite the noted improvements, challenges remain with light utilization efficiency, operational stability, and scalable production. Overall, PSCs provide a viable direction to realize self-powered, miniaturized, and sustainable energy storage for future portable, wearable, and smart electronic devices.

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1. Introduction

1.1 Overview of photo-rechargeable supercapacitors and energy challenges

Supercapacitors with photo-rechargeable capability are potent energy storage devices that have gained significant recognition, especially in smart electronics. They provide a very attractive approach for utilizing sustainable solar energy.[1] Considering the global issues of climate change and environmental pollution, it is necessary to develop and design materials to address energy issues and environmental hazards.[2] The overuse of fossil fuels has caused their continuous depletion and an increase in CO_2 emissions, which are major factors contributing to global warming.[3] Thus, renewable energy sources, including biomass, wind, solar, and ocean energy, are gaining tremendous momentum nowadays.[4] Renewable energy resources can be utilized as the basis of high-performance energy storage devices, which have a minimal impact on global warming.[5] Supercapacitors are known as high-power energy storage devices due to their energy storage characteristics through electrochemical mechanisms.[6]

The rapid charge and discharge, long cycle life (2–3 times that of batteries), high reliability, and high power density (1–2 times that of batteries) of electrochemical capacitors, or supercapacitors, have led to their development as promising devices in recent years.[1] The cycle-life advantage of supercapacitors depends on the specific battery chemistry used for comparison. Supercapacitors demonstrate remarkably longer cycle lifetimes (often exceeding 10^5 to 10^6 cycles) in comparison with many conventional batteries, including lithium-ion batteries, which potentially operate within 500–3000 cycles, depending on the operating conditions and material systems.[7–9]

Recently, scientists have been working on fabricating photo-rechargeable supercapacitors. This facilitates the advanced utilization of renewable light energy.[10] Photo-responsive materials such as ZnO, V_2O_5, TiO_2, and organohalide perovskites are some of the promising elements that are utilized in the fabrication of photo-rechargeable supercapacitors.[11] A hybrid approach combining a solar cell and a supercapacitor has been previously reported, where the solar cell supplies energy to charge the supercapacitor as an integrated energy source.[12] In an integrated study of a polypropylene roll-based supercapacitor and perovskite-based solar cell, their power-pack assembly demonstrated a solar cell efficiency of 10%, which is

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notably higher than most other hybrid systems.[1,13] However, the device size ratio between the supercapacitor and the solar cell remains high. Therefore, to address this issue, self-rechargeable supercapacitors have garnered significant attention.

1.2 Fabrication techniques for high-performance photo-supercapacitors

Distinct types of compounds, such as transition metal oxides/hydroxides, carbonaceous compounds, and conductive polymers, have been explored as supercapacitor electrodes.[13] Binary metal oxides/hydroxides have demonstrated superior electrochemical properties among transition metal oxides/hydroxides compared to single transition metal oxides/hydroxides owing to their high electrical conductivity and multiple oxidation states.[14,15] To date, distinct layered double hydroxide (LDH) nanostructures and binary metal oxides/hydroxides have been documented as promising electroactive materials for capacitors.[16] Among these elements, NiCo oxide/hydroxide-based nanostructures have been identified as appealing electroactive materials due to their various advantages, including high redox activity, flexible ion-exchangeability, low cost, environmental friendliness, and earth abundance.[1]

Materials with higher oxidation states play a significant role in increasing the electrochemical performance of supercapacitors. Furthermore, these materials feature multiple redox-active sites that facilitate reversible faradaic reactions during charge–discharge. In cerium selenide-based electrodes, the presence of cerium in variable oxidation states leads to higher electrochemical activity and improved electron transfer kinetics. This behavior, in turn, enhances the charge storage capacity and energy density. Importantly, materials with a high oxidation state enhance electrical conductivity and ionic diffusion within the electrode structure, thereby promoting stable operation even under mechanical deformation. Lastly, the use of materials with higher oxidation states is a promising approach for creating solid-state, flexible, and high-performance supercapacitors suitable for wearable and portable electronic devices.[17]

Ni and Co ions possess similar potential windows. The co-existence of Ni and Co ions also offers numerous redox reactions within electrodes, which enhances their electrochemical activity.[18] On the other hand, CoOOH possesses higher conductivity, which improves the overall conductivity of the active material during electrochemical processes.[19] NiCo hydroxide/oxide nanosheets (NSs) possess promising electrochemical properties compared to other nanostructures, given their high specific surface area and interconnected arrangement, which facilitate efficient contact with electrolyte ions for reversible and fast faradaic reactions.[15,16] Various growth methods, for instance, electrodeposition, hydrothermal synthesis, chemical bath deposition, solvothermal methods, and microwave-assisted synthesis, can be utilized to develop NiCo nanostructures.[20]

Despite the significance of all these methods, the electrodeposition process is particularly appealing, as it reduces the need for multiple conductive substrates and can be used to deposit nanostructures at low temperatures within a short growth period.[21] Additionally, TiO_2 nanostructures have long been investigated owing to their superior physical and chemical properties.[22] In particular, highly ordered one-dimensional nanostructured TiO_2 nanotubes (TNs) can provide continuous pathways for electron transfer along the nanotube axis, reducing the electrolyte ion transfer distance throughout the nanotube framework.[19,20] The exceptional nanostructure of TiO_2 nanotubes (TNs) also facilitates efficient pathways for ion diffusion and charge transport.[23,24] TNs have recently been explored as effective electrode scaffolds that can enhance the electrochemical properties of various nanomaterials in supercapacitors, photocatalysis, sensors, lithium-ion batteries, and photovoltaic cells. Currently, several methods have been employed for the preparation of TNs, such as hydrothermal/solvothermal methods, template-assisted methods, anodization, and sol–gel methods.[1]

Titanium anodization for the preparation of TNs has several advantages over the abovementioned methods. For example, anodic TNs can be grown vertically on a Ti substrate.[25] Unlike powdered active compounds, they can be directly applied as supercapacitor electrodes without binders given that the Ti metal substrate can serve as a current collector. However, TNs generally have disadvantages, including a low specific capacitance (<1 mF cm^{-2}) owing to their wide band gap (3.2 eV), low electrochemical activity, and poor conductivity (10^{-9} S cm^{-1}). Consequently, several approaches, including modification with nitrogen, metal ions, or graphene; annealing in an H_2 atmosphere; doping; electrochemical reduction; decoration; electrodeposition; and loading, have been employed to address these issues.[1]

Additionally, fabrication techniques play a notable role in determining the electrochemical and photoelectrochemical performance of photo-supercapacitors.[26] The most advanced fabrication principles rely on maximizing charge-transport pathways, light absorption, interfacial contact, and electrode morphology.[27] Among these approaches, *in situ* and binder-free growth strategies are the most useful because they reduce internal resistance and enhance electrical conductivity without inhibiting active surface sites.[28]

Furthermore, electrodeposition remains one of the most fruitful fabrication methods because of its precise control over morphology, composition, and thickness, enabling the formation of uniform nanostructured photoelectrodes directly on conductive substrates.[29]

In addition, this approach is especially efficient for fabricating heterostructure photoelectrodes with NiCo-based compounds deposited on a conductive carbon platform or TiO_2 nanotubes, which can promote a high photoinduced charge separation rate and minimize recombination losses.[30]

Crystalline and hierarchically porous nanostructures with high light-harvesting properties are commonly produced *via* solvothermal and hydrothermal methods.[31,32] Moreover, these methods enable the formation of complex architectures, including hollow structures, nanosheets, and nanoflowers, which can enhance electrolyte accessibility while reducing ion diffusion pathways. Moreover, sol–gel and chemical bath deposition techniques offer scalable and low-cost pathways to

prepare photoactive thin films with tailorable electrochemical and optical characteristics.[33,34]

Lastly, recent progress has also aimed to incorporate all-in-one device fabrication approaches, integrating energy storage electrodes and photoelectrodes into a single platform. Besides, they enhance structural stability and photo-charging performance, and can therefore be used in wearable and self-powered energy systems. Scalable, flexible, and high-performance photo-supercapacitors can also be achieved through precise control over fabrication processes.

1.3 Aim and scope of this review

This review has converged on a critical objective: to provide a holistic understanding of photo-assisted rechargeable supercapacitors (PSCs) as a new category of hybrid energy storage systems that integrate electrochemical storage and light-harvesting functions. This review describes recent developments in photoactive and photothermal nanomaterials, their critical role in improving the performance, conversion efficiency, and stability of PSCs, and the methods used for their production. The fundamental mechanisms underlying photo-induced charge generation, storage, and transfer, and the insights derived from material–structure–property correlations, are highlighted as the focus of this review.

The scope of the research spans both material design and device architecture, aiming to implement transition-metal hydroxides/oxides (*e.g.*, based on NiCo nanostructures), TiO_2 nanotubes, and carbonaceous materials to enable cyclic stability and high energy density. Electrochemical and morphological characteristics of electrodes are influenced by the fabrication methods, including electrodeposition, anodization, and hydrothermal synthesis, reviewed in this study. Conversely, interface engineering barriers, low photo-response efficiency, and scalability have been discussed in detail. The study has provided insights for future research on self-rechargeable, environmentally friendly, and high-performance supercapacitor mechanisms. This can promote the development of new-generation portable and smart energy equipment.

In recent research, it has been shown that there are significant improvements in photo-assisted and solar-rechargeable supercapacitors, with a focus on combining light-harvesting and energy storage capabilities.[35] Remarkable innovations in solar-rechargeable and photo-assisted rechargeable supercapacitors have been demonstrated in existing studies, which have mostly focused on unifying energy storage and light-harvesting functionalities.[36]

This review demonstrates recent developments in photo-assisted asymmetric supercapacitors, which feature embedded $CuCo_2S_4$ and $ZnCo_2O_4$ electrodes, exhibiting improved energy density and capacitance upon surface decoration, owing to the formation of effective charge-transfer systems.[37] Researchers have observed that microsphere-structured $MnO–Mn_3O_4$/carbon composite electrodes exhibit a light-to-charge conversion efficiency of 21.6%.[38]

Indoor photo-supercapacitor operation requires materials with strong visible-light absorption and narrower band gaps, since artificial lighting (*e.g.*, fluorescent lamps, LEDs) is spectrally narrower and weaker than sunlight.[39] As a result, materials need to exhibit low recombination rates, efficient carrier separation, and higher absorption coefficients to maintain photo-charging efficiency under low-intensity indoor illumination, compared to broadband AM1.5G solar conditions.

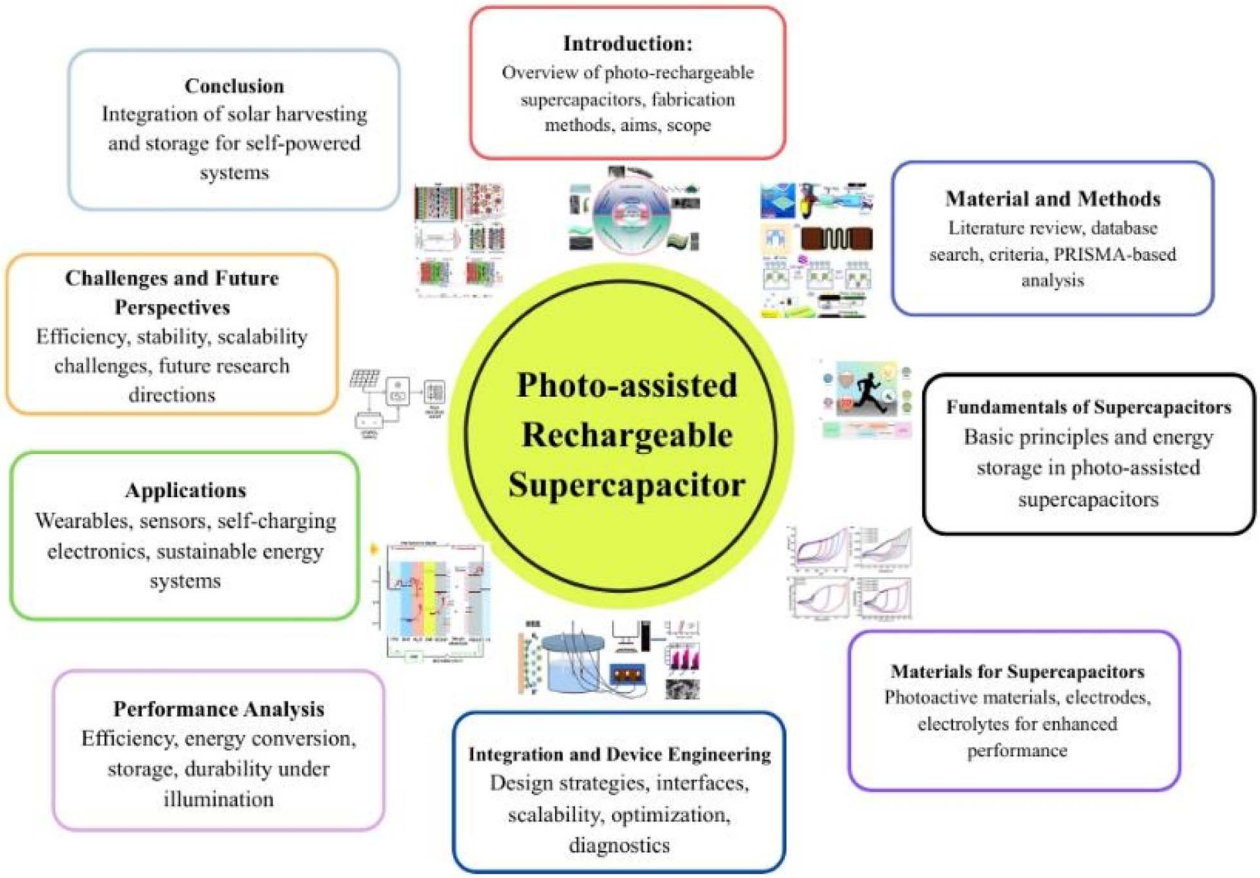
This has led to a notable increase in interest in indoor light-driven storage systems. Emerging reviews, on the other hand, have investigated the effectiveness of photoactive perovskite-based hybrid energy devices. They can provide enhanced photoresponsivity but face challenges with respect to interface degradation and long-term stability.[40] Overall, the existing results indicate a substantial shift toward all-in-one, unified device architectures, in which electrochemical processes and photoconversion synergistically enhance the overall energy storage efficiency.

This review has been made possible through the structural interpretation of peer-reviewed patents, review articles, and other works, which have also helped identify research gaps, highlight other interesting innovations in device architectures and materials, and outline the key issues that have hindered commercialization. The next part of this review is structured as follows: Section 2 covers the fundamentals of photo-assisted rechargeable supercapacitors, and Section 3 examines the materials framework. In addition, the 4th and the subsequent sections address performance, practical applications, and integration, and conclude with the prospects of emerging energy storage technology. The current review article is briefly summarised in Scheme 1.

2. Fundamentals of photo-assisted rechargeable supercapacitors

The development of photo-assisted rechargeable supercapacitors (PSCs) has great potential for both increased energy storage and self-charging capabilities.[41] Photo-rechargeable energy storage devices have opened new avenues for the direct use of solar energy; therefore, the design and assembly of photo-assisted rechargeable supercapacitors for efficient solar energy storage is gaining greater attention. Also, photo-assisted flexible energy storage systems include solar conversion and electrochemical storage. Recent studies have demonstrated high performance, with a maximum energy density of about 60.9 Wh kg^{-1}, stable cycling, and improved capacitance, enabling applications in self-powered and wearable systems.[42] Moreover, photo-assisted rechargeable batteries enable the storage and conversion of direct solar energy, offering a promising pathway for green energy systems. Recent studies indicate improved photoelectrode structure, energy efficiency, and charge transfer, but there are still issues with stability, scale, integration, and conversion efficiency in practical use.[43]

Temperature effects play a significant role in the performance of PSCs under illumination due to photothermal contributions.[44] Additionally, light absorption generates localized heating, which influences ion mobility, reaction kinetics, and electrolyte conductivity.[45] Moderate temperature increases



Scheme 1 Overview of the contents of the current review article.

can enhance charge transport, whereas excessive heating can accelerate the degradation of interfaces, electrolytes, and photoactive materials, thereby reducing long-term efficiency and device stability.[46]

Researchers[41] have developed a notable photoelectrode-assisted asymmetric supercapacitor (ASC), assembled in a particular manner. Nanoflower-like $ZnCo_2O_4$ (ZCO NF) has been designed as a positive electrode that can contribute a higher specific capacitance (563 F g^{-1} at 1 A g^{-1}) under light illumination compared to dark conditions (456 F g^{-1} at 1 A g^{-1}). In line with this, hollow-sphere-structured $CuCo_2S_4$ (CCS HS) has been utilized as the negative electrode,[47] leading to the development of a complete ASC infrastructure capable of efficient photoinduced charge storage. Together, these studies highlight the strategic design of electrode materials with tailored nanostructures to maximize light-assisted electrochemical performance in supercapacitors.

Fig. 1(a) illustrates the layered structure of a photo-assisted supercapacitor, consisting of an FTO substrate, a polyaniline layer, electrolyte, $Co(OH)_2$/$Ni(OH)_2$ electrodes, and a $WTiO_2$ photoactive layer. When the $WTiO_2$ layer is subjected to sunlight, the photons are absorbed by the layer, creating electron–hole pairs (e^-/h^+). These charge carriers move across the electrode layers, leading to redox reactions in the $Co(OH)_2$/$Ni(OH)_2$ electrodes and enabling simultaneous energy harvesting and storage. Fig. 1(b) shows the voltage-time characteristic of the supercapacitor during photo-charging and subsequent discharging at different current densities ranging from 0.04 to 0.14 mA cm^{-2}. The red curve shows the voltage increase under illumination, and the remaining-coloured curves show the discharge profiles at increasing current densities. The higher the current density, the faster the discharge, and light-assisted charging can store more energy than charging without light. Fig. 1(c) compares the photo-charging behavior under the conditions of constant light, darkness, and light-then-dark mode. The blue curve represents photo-charging in the light, the black curve shows charging in the dark, and light-then-dark charging is shown in red. This study has demonstrated an increase in energy storage due to photon-induced charge generation and has indicated that PSCs can successfully utilize stored electrochemical energy for direct solar energy conversion.

Table 1 summarises recent studies on PSCs, highlighting the diversity of device architectures, from asymmetric and hybrid designs to all-in-one integrated systems. The instances above provide an account of the extent to which the explored strategies can improve charge transfer, overall energy storage efficiency, and light harvesting.

Beyond asymmetric ASCs, Arif *et al.* reported the fabrication of binder-free three-dimensional $NiCo_2S_4$@Cu_2O@CF nanowire

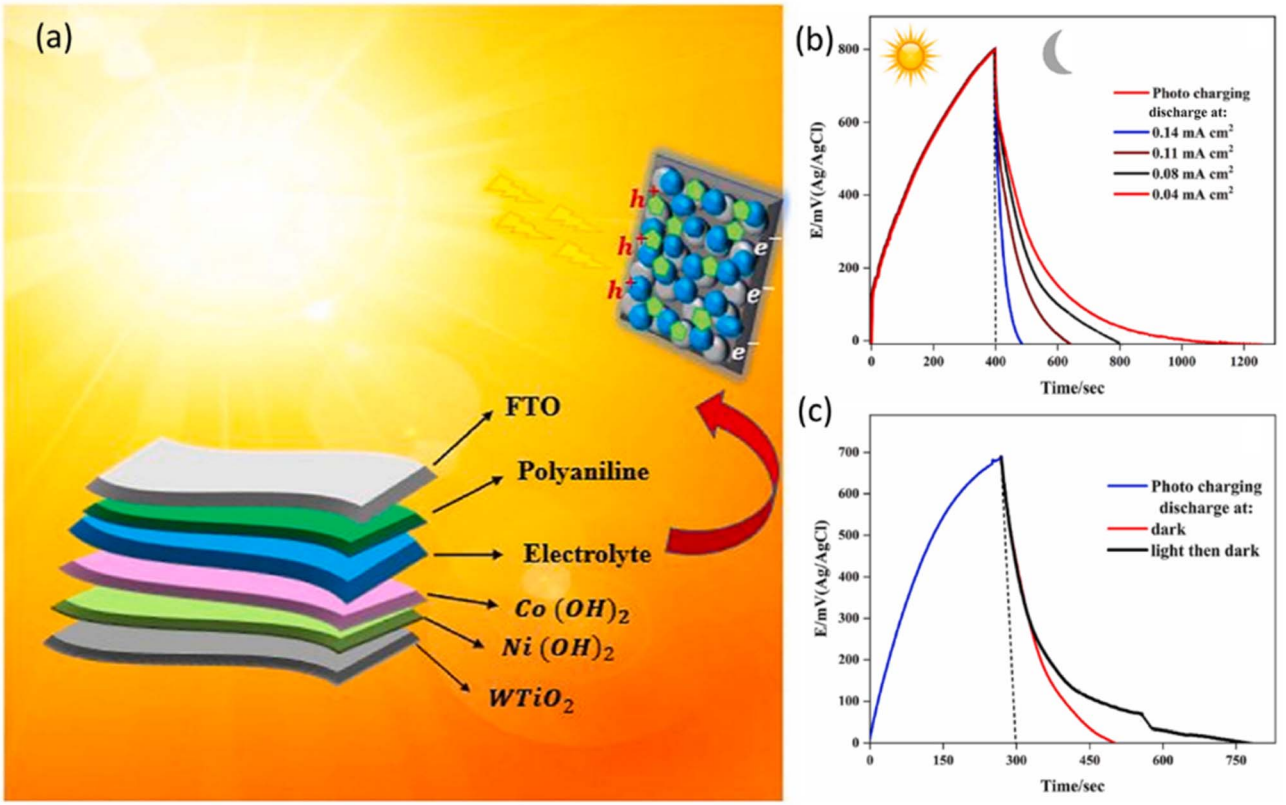


Fig. 1 Photo-assisted or solar-rechargeable supercapacitor system: (a) device architecture, (b) photo-charging and discharge performance at different current densities, and (c) light *vs.* dark charge–discharge comparison. Reproduced with permission from ref. 1. Copyright 2022, Elsevier B.V.

Table 1 Analysis of recent literature on photo-assisted rechargeable supercapacitors

Author(s) (year)	Focus/main theme	Cycling stability	Photo-charging/ conversion efficiency	Specific capacitance	Reference no.
Dong *et al.* (2023)	Light-assisted energy storage principles and perspectives	—	—	—	48
Arif *et al.* (2025)	$NiCo_2S_4$@Cu_2O nanowire heterostructures	∼90% retention after 5000 cycles	∼128.5% coulombic efficiency under illumination	∼1620 F g^{-1} (reported electrode performance)	41
Bhattacharjee *et al.* (2025)	A comprehensive review on integrated photo rechargeable batteries- supercapacitors	—	—	—	49
Namsheer and Rout (2021)	Hybrid photo-supercapacitors review	—	—	—	50
Zhao *et al.* (2023)	Photo-assisted asymmetric supercapacitors	∼85–90% retention after ∼5000 cycles	Significant photo-enhanced charge storage under illumination	∼694 F g^{-1} (photo-electrode system)	47
Tuc Altaf *et al.* (2023)	All-in-one integrated photo-supercapacitors	—	Solar-to-storage efficiency up to ∼20% summarized	—	51
Martinez *et al.* (2023)	Systematic review framework (energy materials)	—	—	—	52
Momeni *et al.* (2023)	Photo-assisted rechargeable supercapacitors based on nickel–cobalt-deposited tungsten-doped titania photoelectrodes	∼94% retention after 10 000 cycles	Photo-enhanced capacitance increases under light	∼955.6 mF cm^{-2} under illumination	1

heterostructures on copper foam as bifunctional photoelectrodes.[41] Under illumination, the contact potential difference increased from 65 mV to 138 mV, while the charge–discharge capacitance improved from 733.3 to 1156.7 mF cm^{-2} at 20 mA cm^{-2}. Under illumination, the coulombic efficiency was 128.52%, indicating effective photo-induced charge generation. In addition, in photo-assisted energy storage systems, apparent efficiencies greater than 100% are possible due to the presence of other photo-generated charge carriers that add to the discharge capacity upon illumination.[53] Moreover, light irradiation in these situations creates additional holes and electrons in the photoactive electrode, which are involved in electrochemical reactions and augment the discharge capacity measured under electrochemical conditions compared to that due solely to electrochemically stored charge.[54] The PSC was extremely stable and exhibited electrochemical stability for more than 5000 cycles, demonstrating that it could be used as a long-term, highly sensitive light-driven energy storage device.[41]

Fig. 2(a) depicts the fabrication of Cu_2O@CF and NCS@Cu_2O@CF photo-assisted supercapacitor electrodes, starting with copper foam (CF), followed by the growth of $Cu(OH)_2$ nanowires, which are annealed to form Cu_2O@CF. Subsequently, NCS is electrochemically deposited to obtain the NCS@Cu_2O@CF structure. Fig. 2(b) illustrates the specific capacitance retention of 3D $NiCo_2S_4$@Cu_2O nanowires supported on copper foam over 5000 charge–discharge cycles. The device maintains excellent stability, retaining 85.0% (light) and 85.8% (dark) of its initial capacitance, confirming its superior durability and consistent electrochemical performance under both illumination and dark conditions. The inset shows an SEM image of the nanowire morphology.

Inexhaustible solar energy has now been recognized as the most fundamental substitute for conventional fossil energy. Solar energy can be captured with solar cells, converted to electrical power, and later stored in batteries or supercapacitors. To achieve higher energy utilization efficiency, integrating solar energy storage and conversion has become increasingly significant. Recent studies have shown that a photo-assisted rechargeable supercapacitor, integrating a working photoelectrode and counter electrode, can form a nanoporous Cu@Cu_2O (NPC@Cu_2O) hybrid array electrode, thereby enhancing the specific charge capacity. The novel nanoporous/array hybrid structure ensures efficient light illumination, enhancing the utilization of solar energy. It also delivers a specific capacitance of 782 F g^{-1} at 1 A g^{-1} under illumination, which is 37.9% higher than in the dark.[55]

Kinetic analysis and capacity-enhancing mechanisms have shown that the holes produced on the surface of Cu_2O under light serve as additional active sites that enable proton insertion into the facets of Cu_2O. These findings have therefore opened a pathway towards the direct storage of large amounts of solar energy. The basic principle of PSCs is that light photons are captured in photoactive electrodes and electron–hole pairs are produced, which contribute to charge separation and redox reactions in the electrochemical system.[56]

2.1 Photoelectrochemical mechanism

Photo-assisted rechargeable supercapacitors operate on the principle that the generation and separation of photogenerated charge carriers at the semiconductor photoelectrode control its operation. When electrons are excited to the conduction band by photons possessing sufficient energy from light irradiation, the electrons and holes combine to produce electron–hole pairs:

Photoexcitation

$$\text{Semiconductor} + \hbar\nu \rightarrow e^- + h^+$$

The photon energy must be equal to or greater than the bandgap energy of the semiconductor:

$$E_g = \hbar\nu$$

Following excitation, efficient charge separation occurs, where electrons migrate toward the conduction band and are transferred to the external circuit, while holes move toward the electrode–electrolyte interface to participate in oxidation reactions:

$$e_{CB}^- \rightarrow \text{external circuit}$$

$$h_{VB}^+ \rightarrow \text{electrolyte interface}$$

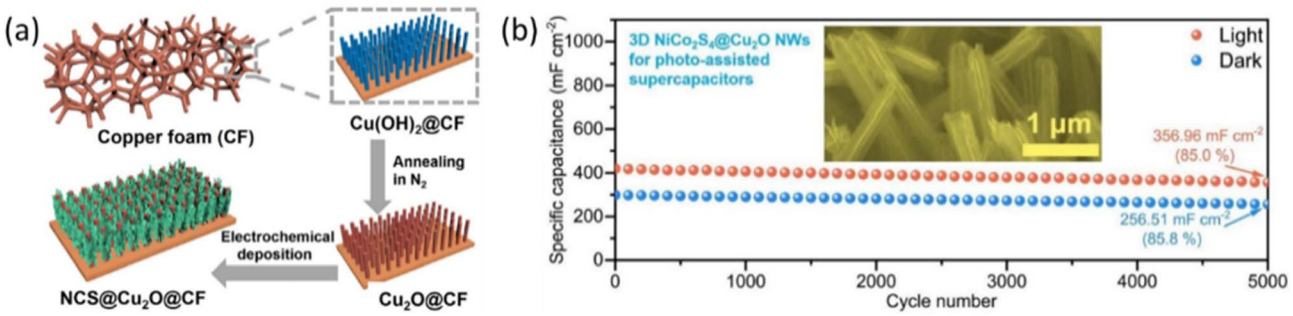


Fig. 2 (a) Steps in the synthesis of NCS@Cu_2O@CF NW heterostructures. (b) Cycling stability of 3D $NiCo_2S_4$@Cu_2O nanowire-based photo-assisted rechargeable supercapacitors under light and dark conditions. Reproduced with permission from ref. 41. Copyright 2025, Elsevier.

The asymmetric, integrated all-in-one and tandem designs are key device architectures, optimized using nanostructured electrodes to enhance light absorption and reducing recombination.[57] On the other hand, $ZnCo_2O_4$, $NiCo_2S_4$, TiO_2, and $CuCo_2S_4$ are key materials that play a fundamental role in facilitating charge transport and light harvesting. Density functional theory (DFT) is one of the most extensively used methods for investigating charge transfer systems. This has demonstrated the effectiveness of photogenerated carriers in ion extraction/insertion.[58] Nevertheless, PSCs face challenges such as low cycling stability, operational degradation, and low photo-charging efficiency, despite their immense advantages. This has led to the continuous investigation of hybrid nanostructures, improved device designs, and more practical, off-grid, self-powered materials and systems.[59]

In PSCs, the incident photon-to-current conversion efficiency (IPCE) is an important parameter that quantifies the fraction of incident photons that generate usable charge carriers in the photoelectrode. Additionally, the IPCE typically varies strongly with wavelength because semiconductor photoelectrodes absorb light based on their optical properties and bandgap. For example, wide-bandgap materials, such as ZnO and TiO_2, mainly absorb in the ultraviolet region, whereas heterostructures or narrow-bandgap materials extend absorption into the visible spectrum. As a result, dye-sensitization strategies and heterojunction engineering are often applied to increase the spectral response as well as broaden photon harvesting in PSC systems.[60]

The incident photon-to-current conversion efficiency (IPCE) can be mathematically expressed as follows:

$$\text{IPCE}\ \ (\%) = \frac{1240 \times I}{\lambda \times P_{\text{light}}} \times 100$$

This parameter quantifies the efficiency with which incident photons are converted into usable electrical charge carriers in photo-assisted supercapacitor systems.

The internal quantum efficiency (IQE) quantifies the fraction of absorbed photons that ultimately leads to stored electrochemical charge. Additionally, in PSCs, this process involves various steps, such as photon absorption, exciton generation, and charge transport and separation in the semiconductor, as well as the participation of charge carriers in electrochemical redox reactions at the electrolyte–electrode interface. Furthermore, losses can occur due to inefficient carrier injection or recombination in the storage layer. Hence, conductive scaffolds, heterojunction interfaces, and nanostructured electrodes are widely employed to improve the carrier separation efficiency and maximize the conversion of photogenerated carriers into stored electrochemical energy.[61]

However, many early PSC research studies evaluated performance under indoor light sources or monochromatic illumination, which do not accurately reflect real-world operating conditions. Hence, recent research increasingly reports device performance under standard AM1.5G solar illumination to offer realistic comparisons. Moreover, under these conditions, integrated photo-capacitor devices have demonstrated improved performance because of the better matching between electrochemical storage processes and photovoltaic conversion.[62]

Current studies report remarkable progress in the overall solar-to-stored-energy conversion efficiency of photo-assisted supercapacitor systems. Additionally, integrated photo-capacitor devices combining photovoltaic units with supercapacitors have achieved conversion-storage efficiencies exceeding ~20% in optimized architectures, including perovskite-solar-cell–supercapacitor hybrids.[63]

Subsequently, the practical efficiency of most PSC systems typically remains below ~10–15% under AM1.5G illumination, primarily due to losses from photon absorption mismatch, interfacial resistance, charge recombination, and energy transfer between the storage units and the photoconversion component. Moreover, from a theoretical perspective, the upper limit is constrained by the photovoltaic conversion efficiency of the photoactive material (*e.g.*, Shockley–Queisser limit for single-junction semiconductors is ~33%) combined with electrochemical storage efficiency.[64] Consequently, the theoretical limits for incorporated PSC systems are generally estimated to be below ~25–30% overall solar-to-stored energy efficiency, depending on the material systems and device architecture.

3. Materials for photo-assisted rechargeable supercapacitors

The performance of photo-assisted rechargeable supercapacitors greatly depends on the synchronization among electrode composition, electrolyte design, and photoactive materials.[47,50] Optimizing these elements has enhanced charge separation, light absorption, overall energy storage efficiency, and ionic transport. Therefore, superior photoelectrochemical performance has enabled extended stability under cycling conditions and continuous improvement.

3.1 Photoactive materials

The increasing development of photo-induced methodologies for various energy conversion and storage processes, as well as for photocatalytic processes, such as oxygen evolution reaction (OER), hydrogen evolution reaction (HER), rechargeable batteries, and oxygen reduction reaction (ORR), is beneficial due to their low cost, ready accessibility, and environmental friendliness. In addition, photo-supercapacitors incorporate photovoltaic energy conversion with electrochemical storage. Applying photoactive materials, including dyes, polymers, and metal oxides, remarkably improves capacitance and energy conversion efficiency, and facilitates promising applications in wearable devices.[65]

Photoirradiation-enhanced capacitance, also known as photo-assisted rechargeable supercapacitors, has emerged as a promising energy storage approach.[66] This type of device features photoactive properties integrated into a usable capacitive electrode material, thereby accelerating the charging

process or potentially enhancing photothermal or photoconductive effects.[67] Additionally, interface engineering of heterostructure materials plays a significant role in increasing the transport and separation of photo-generated charge carriers at the electrode interface. This improved charge separation effectively minimizes recombination losses and significantly increases the energy storage capacity and electrochemical performance of PSCs.[68] Hence, enhanced interfacial contact within the electrode and improved photoactive properties through nanostructuring are promising strategies to achieve a higher performance. Improved interfacial contact and nanoscale structuring are conducive to achieving an optimal band gap, which facilitates the transfer of photo-generated carriers. Thus, an investigation into novel combinations of photoactive and electrode materials is necessary at this stage to better understand the structure–property relationships of these heterostructure materials.

Heterogeneous photocatalysts, as well as photoelectrochemistry, are important approaches for converting solar energy into chemical fuels, often referred to as solar fuels.[69] Furthermore, it is difficult to design efficient photoactive materials due to the trade-off between visible-light absorption and photoactivity. Wide-bandgap semiconductors provide strong redox driving forces but absorb primarily ultraviolet light, limiting their use of solar energy. In contrast, narrow-bandgap semiconductors are visible-light absorbers but lack sufficient energy to drive oxidation and reduction reactions in the photocatalytic cycle. Researchers can modify wide-bandgap materials or build heterostructured composites of wide- and narrow-bandgap semiconductors. These strategies enhance spectral sensitivity and photocatalytic activity and promote efficient charge separation.[69]

In the study by Najafi *et al.*,[70] they fabricated several potential CS@TNT, NCS@TNT, and NS@TNT photoactive electrodes for photo-rechargeable supercapacitors. In photoelectrochemical measurements, the NCS@TNT-1 sample demonstrated higher ability to separate and generate photogenerated charges than the bare NS@TNT, CS@TNT, and TNTs. The NCS@TNT-1 electrode exhibited the highest capacity of over 471.6 mF cm^{-2} (at 0.7 mA cm^{-2}), which is about 11-times greater than that of bare TNTs (44 mF cm^{-2}). The specific capacitance of the NCS@TNT-1 electrode (as the best sample) increased by approximately twofold to 955.6 mF cm^{-2} upon light illumination. Three photo-chargeable asymmetric supercapacitors were fabricated using PVA–KOH as the separator/electrolyte, and NCS@TNT as the electrode. The specific capacitance of the fabricated supercapacitor increased by 1.57 times when illuminated. The device demonstrated exceptional stability over 10 000 discharge and galvanostatic charge cycles, with capacity retention of 94% and 87% under light and dark conditions, respectively.[70] Under illumination, the discharge time also significantly increased.

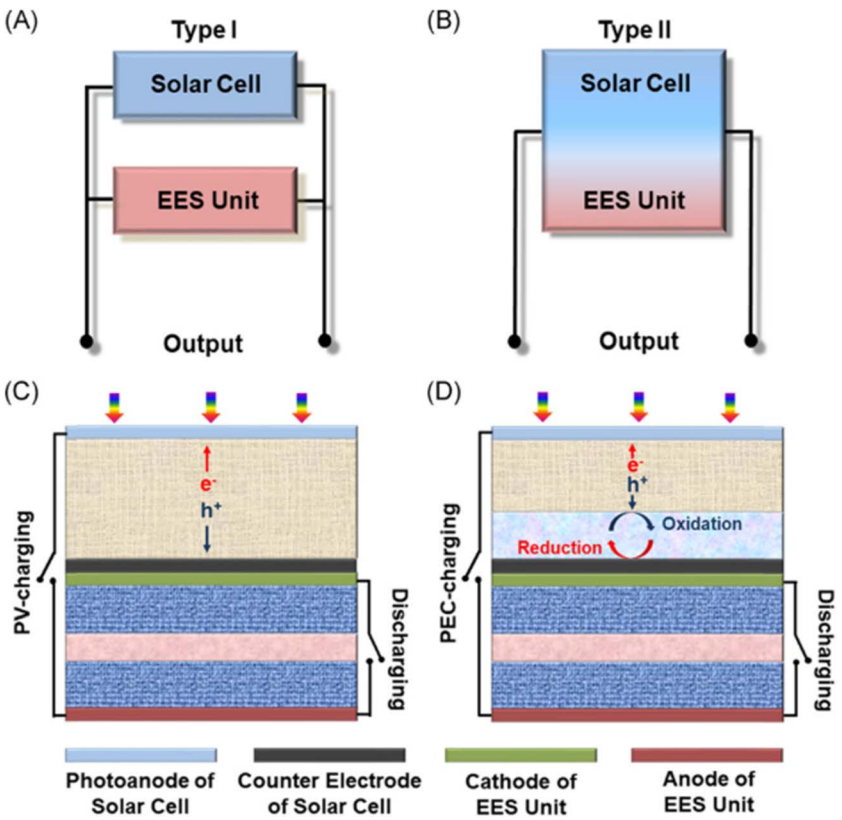


Fig. 3 Schematic of integration modes in photo-charging hybrid devices: (A) type I and (B) type II configurations. Photo-charging and discharging mechanisms of (C) PV-rechargeable and (D) PEC-rechargeable integrated devices. EES denotes electrochemical energy storage; PEC, photoelectrochemical; and PV, photovoltaic. Reproduced from ref. 10 from Wiley-VCH. Reproduced with permission from ref. 10. Copyright 2021, Wenzhou University and John Wiley & Sons Australia, Ltd.

Between the two modes mentioned above (Fig. 3), type I (Fig. 3(A)) represents the simplest connection method and can be applied to fabricate a wide range of EES devices and solar cells. Similarly, Fig. 3(B) depicts a type II system that potentially incorporates EES devices and solar cells more closely, forming a hybrid or monolithic structure. This configuration enables direct charge transfer between the two units, potentially improving storage efficiency and energy conversion. Additionally, Fig. 3(C) represents the photovoltaic (PV)-charging mechanism of type-I systems. Moreover, when illuminated, electron–hole pairs are generated by the photoanode of the solar cells. Subsequently, the electrons (e^-) move toward the counter electrode, while holes (h^+) migrate in the opposite direction, allowing the EES unit to store energy during the charging phase. Afterwards, during discharge, the EES unit transfers the stored energy to the external circuit. Additionally, Fig. 3(D) shows the photoelectrochemical (PEC) charging system, which is typical of Type II systems. Here, light absorption initiates concomitant reduction and oxidation reactions at the photoelectrode interfaces. The resulting electrons and holes undergo redox reactions at the anode and cathode of the EES unit, enabling effective energy storage. The chemical energy stored during charging can be converted back into electrical energy during discharge (Table 2).

Photo-assisted rechargeable supercapacitors, particularly NCS@TNT-based electrodes, demonstrate significantly enhanced charge generation, separation, and storage under light illumination. Improved interfacial engineering and nanostructuring optimize photocarrier transfer, resulting in higher capacitance, extended discharge times, and excellent cycling stability, highlighting their potential for efficient, sustainable, and portable solar energy storage applications.

In addition to traditional sulfides and metal oxides, new hybrid photoactive materials and semiconductor heterostructures are attracting increasing interest for PSC applications.[71] Also, the strategic interconnection of wide-bandgap semiconductors with narrow-bandgap materials enables broader light absorption and enhances photocarrier lifetime and mobility.[72] Specifically, p–n heterojunctions and Z-scheme structures have been shown to be useful for suppressing electron–hole recombination and enabling directional charge transfer.[73] Moreover, the use of conductive scaffolds, such as graphene or carbon nanotubes, further enhances mechanical stability and electrical conductivity.[74] Localized energy states created by doping strategies and surface defect engineering accelerate redox kinetics and enable the absorption of visible light.[75] After that, these material innovations, in addition to improving photo-charging efficiency, also enhance long-term cycling stability and high-rate performance. In turn, hybridization and heterojunction engineering continue to be important contributors to high-performance photo-supercapacitors[76] for indoor lighting or low-intensity operation, making them more relevant to distributed energy systems and wearable electronics (Table 3).

3.2 Electrode materials

The key properties and dynamic applications of the platforms have been realised by the photo-rechargeable energy storage modules. In particular, the intermittent nature of solar energy demands a combination of different types of storage systems, primarily supercapacitors (SCs) and batteries.[76] Photo-sensitive electrodes allow the enhancement of electrochemical properties when the electrode is used in large-area photo-rechargeable energy storage systems under light illumination.[77,78] However, these approaches face some limitations and significant challenges. To overcome the energy losses incurred during energy transportation between energy storage devices and solar cell modules, recent research has focused on the development of single devices that integrate both energy storage functions and light collection.[50,79] To further address the limitations associated with interlinking the modules of photovoltaic cells with energy storage components, recent research has focused on creating single devices that combine both light-harvesting and storage devices.[50]

Liu *et al.*[80] investigated heavy mass-loading electrodes for SCs and secondary batteries. They explored the effects of heavy mass loading on advanced secondary batteries, including lithium-ion batteries (LIBs), sodium-ion batteries (SIBs), lithium-air batteries (LABs), magnesium-ion batteries, zinc-ion batteries, calcium-ion batteries, aluminum-ion batteries, potassium-ion batteries, and SCs. LABs were introduced as a substitute for LIBs. Additionally, LABs are sometimes referred to as non-aqueous batteries. LABs operate *via* reactions of Li metal with O_2 from the air, forming lithium peroxide (Li_2O_2) at the surface of the positive electrode during discharge.[81] Furthermore, the reduction of O_2 gas at the cathode surface generates current.

In these devices, particularly in supercapacitors, efforts have been devoted to designing photoactive electrodes with enhanced capacitive performance under illumination. Examples of these strategies include Zn-ion capacitors with a V_2O_5-activated carbon electrode, ZnO-nanoflakes/rGO-based solar-assisted supercapacitors, and photo-assisted asymmetric supercapacitors using $ZnCo_2O_4$ or $CuCo_2S_4$, among others. There are conceptual and practical difficulties associated with

Table 2 Quantitative comparison between type I and type II integration modes in terms of overall efficiency losses

Parameter	Type I integration (PV-charging)	Type II integration (PEC-charging)
Pathway of energy conversion	PV cell → external circuit → supercapacitor	Direct photoelectrode → electrochemical storage
Transfer of charge	Indirect	Direct
Complexity of device	Higher	Lower

Table 3 Literature matrix of photoactive materials for photo-assisted rechargeable supercapacitors

Ref.	Fabrication/structure	Photoactive material/system	Key performance metrics	Device type	Key findings/contribution
68	Interface engineering	Heterostructure photoactive materials	Improved carrier separation	PSC electrode materials	Highlighted the importance of heterostructure interfaces for efficient photocarrier transport
70	Heterostructure on TiO_2 nanotubes	CS@TNT, NCS@TNT, NS@TNT	471.6 mF cm^{-2} (dark); 955.6 mF cm^{-2} (light); 10 000 cycles stability	Photo-rechargeable supercapacitor	NCS@TNT showed superior photogenerated charge separation and ~2× capacitance increase under illumination
50	Nanostructured materials	Hybrid semiconductor composites	Enhanced light absorption	PSC systems	Demonstrated broader spectral response for improved photocharging efficiency
72	Semiconductor heterojunction	p–n junction/Z-scheme architectures	Reduced recombination losses	Photo-assisted devices	Improved directional electron–hole transport and charge storage
74	Conductive composite structures	Graphene/carbon nanotube hybrid scaffolds	Increased conductivity and stability	PSC electrodes	Carbon frameworks enhance electron transport and structural durability
75	Defect-engineered materials	Doped semiconductors	Faster redox kinetics	Photo-supercapacitors	Surface defects and doping improve visible-light absorption and electrochemical activity

these systems, including the limited light absorption, ineffective photoresponse, poor electrochemical functionality, and poor stability.[82] The reduction in energy density is the greatest challenge to their practical implementation for supplying power to advanced electronic equipment. To address this problem, research has primarily focused on the development of electrode materials that possess high electrochemical performance through conventional synthesis approaches, as demonstrated in the case of atomic doping, morphology control, and hybridization with other active substances.[83]

Fig. 4(a) displays a photo-rechargeable supercapacitor device. It utilizes activated charcoal electrodes mounted on graphite sheets within a transparent container (cuvette) filled with an aqueous electrolyte containing the photolabile molecule 2-nitrobenzaldehyde. A solar simulator directs light onto the cell, charging the device by initiating a photochemical reaction within the electrolyte. Copper tape connects the electrodes to an external voltmeter, enabling monitoring of cell voltage. This setup integrates light-harvesting and storage.

In Fig. 4(b), step I involves light absorption by Cu_xO NWs, generating electron–hole pairs. In step II, the generated electrons migrate toward the NiO surface, while holes move outward. The integration of NiO provides additional pseudocapacitance, resulting in higher capacitance and photocurrent density under illumination, as shown in the graphs (Fig. 4(b)). The electrode exhibits a higher specific capacitance under light

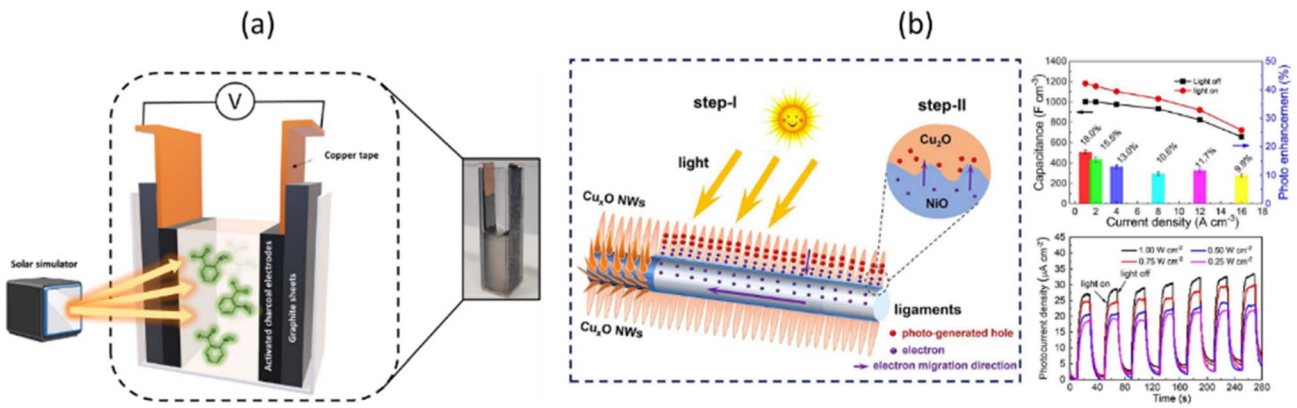


Fig. 4 (a) Photo-rechargeable supercapacitor design. Reproduced with permission from ref. 82. Copyright 2025, Wiley-VCH. (b) Multi-layered porous photoelectrode with dual-component photosensitive (Cu_xO) and pseudocapacitive (NiO) demonstrates enhanced photo-assisted supercapacitor performance. Reproduced with permission from ref. 83. Copyright 2023, Elsevier B.V.

(photo-enhancement) that decreases with increasing current density. The photocurrent density is stable and reversible, increasing with increasing light intensity.

Fig. 5(a) and (b) schematically distinguish between a double-layer capacitor (EDLC) and a pseudocapacitor. In an EDLC (left), ions are stored at the electrode–electrolyte interface *via* physical adsorption/desorption. In contrast, in a pseudocapacitor (right), the storage mechanism is reversible, involving fast faradaic redox reactions at the electrode surface, and is often accompanied by ion intercalation/deintercalation, which gives the capacitor a larger specific capacitance. Fig. 5(c) displays the wide range of two-dimensional (2D) nanomaterial families, such as graphene, transition metal dichalcogenides (TMDs), MXenes, and metal oxides, which have been extensively investigated as superior electrode materials for high-performance supercapacitors due to their high surface area and good conductivity.

To further explain the faradaic charge storage mechanism in transition metal-based electrodes, the redox reactions occurring at the electrode surface can be represented as follows:

$$Ni(OH)_2 + OH^- \leftrightarrow NiOOH + H_2O + e^-$$

$$Co(OH)_2 + OH^- \leftrightarrow CoOOH + H_2O + e^-$$

These reversible redox reactions contribute significantly to pseudocapacitance by enabling rapid electron transfer and ion diffusion at the electrode–electrolyte interface, thereby enhancing the overall energy storage performance of photo-assisted rechargeable supercapacitors.

Recent findings have revealed that light irradiation, particularly sunlight, on photosensitive electrode materials can be a novel approach for enhancing their energy storage capabilities, generating significant interest in the field of photo-assisted or photo-charging energy storage. This new strategy will enable energy production and storage components to be combined into a single device, significantly reducing the form factor of the energy storage system.[83]

The main conclusion is that photo-rechargeable energy storage is one of the key directions, where the detached solar

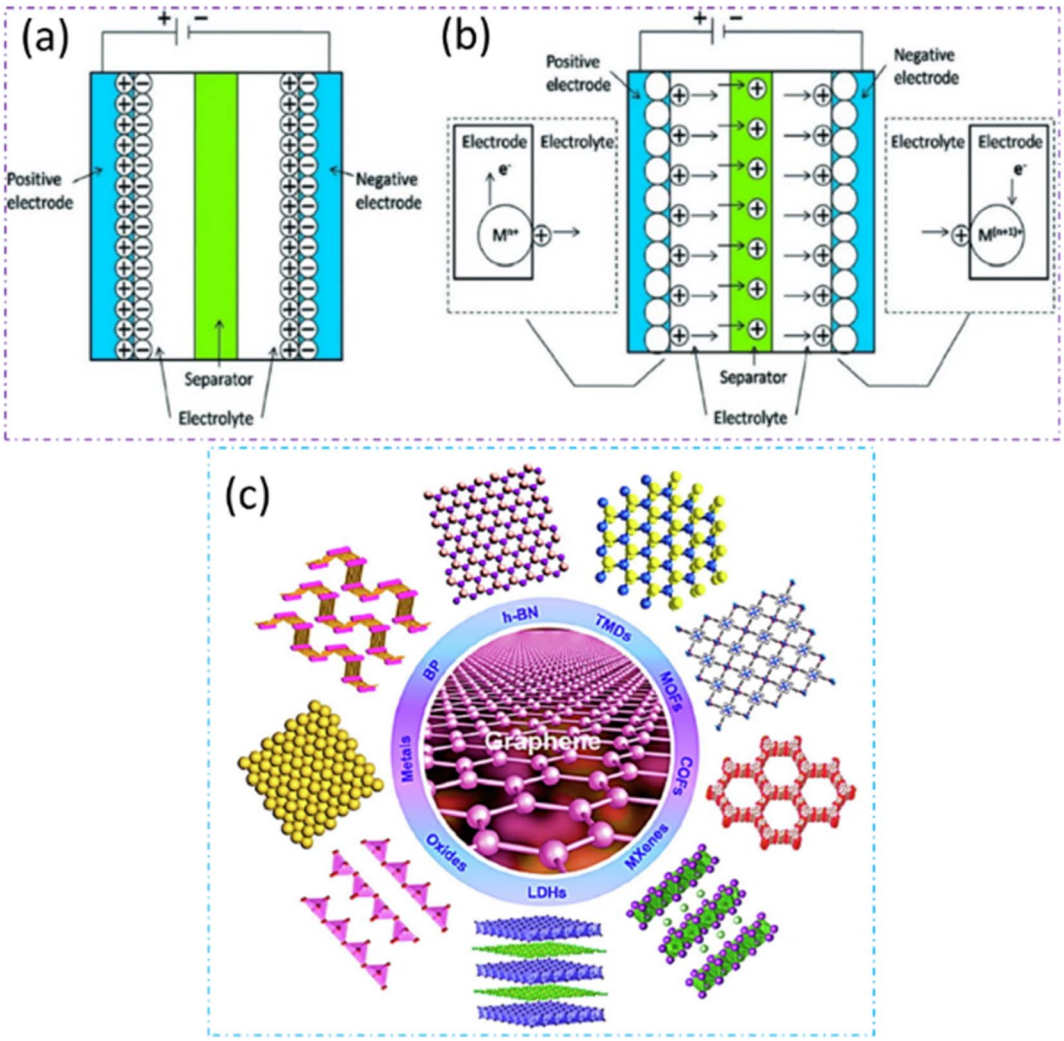


Fig. 5 (a) and (b) Schematic of EDLC and pseudocapacitor. Reproduced with permission from ref. 84. Copyright 2017, Elsevier B.V. (c) Illustration of various types of two-dimensional nanomaterials as electrode materials in supercapacitors. Reproduced with permission from ref. 85. Copyright 2021, Elsevier.

Table 4 Literature matrix of photo-assisted energy storage systems and electrode strategies

Author/ref.	Photoactive/electrode material	Energy storage system	Key performance/role	Device configuration	Limitations/ challenges
76	Supercapacitors + batteries	Hybrid energy storage modules	Addresses the intermittency of solar energy	Integrated renewable energy storage	Requires multiple storage systems
77 and 78	Photo-sensitive electrodes	Photo-rechargeable supercapacitors	Improved electrochemical performance under illumination	Light-responsive electrode systems	Limited efficiency under weak light
79	Photoactive electrodes + storage units	Integrated photo-storage devices	Reduces energy loss during energy transfer	Single-device architecture	Complex integration design
82	ZnO nanoflakes/rGO	Solar-assisted supercapacitors	Enhanced light absorption and charge storage	Hybrid PSC system	Stability and photoresponse issues
83	Doped and hybridized materials	Advanced PSC electrodes	Improved electrochemical performance	Nanostructured electrodes	Complex synthesis routes

cells and storage cells are replaced by integrated devices. This approach targets photo-assisted rechargeable supercapacitors, which utilize photoactive electrodes (*e.g.*, transition metal oxides) to enhance capacity and response to light irradiation, simplify the energy system, and mitigate the low energy density issue (Table 4).

3.3 Electrolytes

An increase in the capacitance of photo-assisted rechargeable zinc-ion hybrid supercapacitors can be achieved by adding bromide ions, in combination with appropriate supporting electrolytes. The most popular aqueous electrolytes include aqueous hydroxides due to their high ionic conductivity and low cost. Electrolytes based on ionic liquids are especially appealing for energy-storage applications at high temperatures due to their low volatility, high thermal stability, non-flammability, and low-pressure vapor properties. Electric double-layer capacitors (EDLCs) store charge through the adsorption of ions at the electrode interfaces, rather than through redox reactions, and as a result, they exhibit superior cycling stability and a long operating lifetime compared to other energy-storage systems.

The performance of EDLCs is affected by electrode surface area, ion size, electrolyte viscosity, ionic conductivity, and other parameters. Given their high surface area compared to other materials, carbon electrode materials dominate within EDLCs. Their capacitance depends on the electrode material's surface area, porous architecture, and carbon composition. EDLC-based supercapacitors have advanced given that they do not undergo phase changes, unlike batteries. At the electrode surface level, several models and theories have been developed to explain the interconnection of electrolytic ions, resulting in the formation of a double layer at the interface, as suggested in a previous study.[86]

Zinc-bromide (Zn_2-Br_2) batteries in aqueous solution are of great interest due to their inherent safety, high theoretical capacity, and high discharge voltage.[55,56] However, the efficacy of bromide conversion hosts is significantly impaired by the polybromide shuttle effect and slow redox processes.[87,88] The cells contain an aqueous zinc bromide ($ZnBr_2$) electrolyte solution that is constantly recirculated through the electrolyte compartments *via* external storage tanks. Additionally, Zn–Br_2 electrolytes and redox-mediated systems are discussed as representative examples of redox-active electrolytes that can increase ionic conductivity and charge storage under illumination. Moreover, these systems offer valuable insights into ion transport mechanisms, redox mediation, and electrolyte design strategies that are also applicable to PSC architectures.

The electrochemical reactions governing the Zn–Br_2 system can be represented as follows:

$$Zn^{2+} + 2e^- \leftrightarrow Zn$$

$$2Br^- \leftrightarrow Br_2 + 2e^-$$

These reversible redox reactions facilitate charge storage *via* ion conversion, thereby enhancing capacitance and improving electrochemical performance in photo-assisted energy storage systems.

As the battery charges, aqueous zinc ions flow across the membrane to the anode/anolyte side and are reduced to metallic zinc, as shown by the above equations. Metallic zinc is directly deposited from the liquid phase onto a solid electrode (carbon-based). Meanwhile, aqueous bromide ions are oxidized to liquid bromide at the cathode/catholyte interface of the cell. An example of these solutions is the use of binary redox species as the electrolyte, which ensures better ionic conductivity and an expanded electrochemical window, as well as enhanced longevity of zinc anodes.[57,59,60] Besides this advantage, these systems also feature an innovative approach for energy conversion and storage devices, including the development of compact, direct-sunlight storage devices using dual-acting electrodes.

Photoactive electrolyte storage under light is a novel and promising concept that can create new opportunities in photo-

charged devices, solar energy collection, and storage. The electrolyte in the device is a small molecule, 2-nitrobenzaldehyde, which undergoes a chemical change upon light irradiation to generate photoacid species. Photoacids exhibit greater dissociation in the photoexcited state than in the ground state.[82]

The practical application of a photoacid–electrolyte supercapacitor is illustrated in Fig. 6(A–C) in three stages. We connected a series of five devices (diagram at bottom left) in the experiment to generate a sufficiently large voltage to activate a light-emitting diode (LED). After the first 3 h photoactivation, the device was charged at 1 A and continued under white light (70 mW cm^{-2}) (first step). As a result, charging was terminated with a white LED linked to the supercapacitors, as shown in Fig. 6(A). In the second step, as shown in Fig. 6(B), the LED was turned on by linking with the assembled devices. Thereafter, LED emission persisted for approximately 2 min after light irradiation was turned off, as shown in the third step of Fig. 6(C); this resulted in a high energy output of the 'photo-rechargeable supercapacitor'. On the other hand, Fig. 6(D) highlights the performance limitations of supercapacitors that rely on conventional electrolytes. Due to their short cycle life, narrow electrochemical window, and low energy density, organic and aqueous electrolytes exhibit a poor performance. In contrast, electrolytes based on ionic liquids deliver a long cycle life, a broad electrochemical window, higher energy density and capacitance, and high thermal stability, highlighting their superiority for high-performance devices.

Photo-rechargeable devices are not immune to safety concerns, particularly regarding the photostability of the electrolytes under continuous illumination. Moreover, long-term light exposure can cause decomposition of the electrolyte, changes in pH, or the production of active species that degrade electrode materials.[89] Therefore, to ensure long-term operational stability, electrochemical stability, and device safety in photo-assisted energy storage systems, chemically stable, photo-inert electrolytes and robust encapsulation methods are required.

Innovations in supercapacitors focus on photo-assisted charging and electrolyte optimization, which are discussed in this section. Utilizing ionic liquids addresses the limitations of aqueous and organic electrolytes by providing greater stability, wider electrochemical windows, and higher energy density.

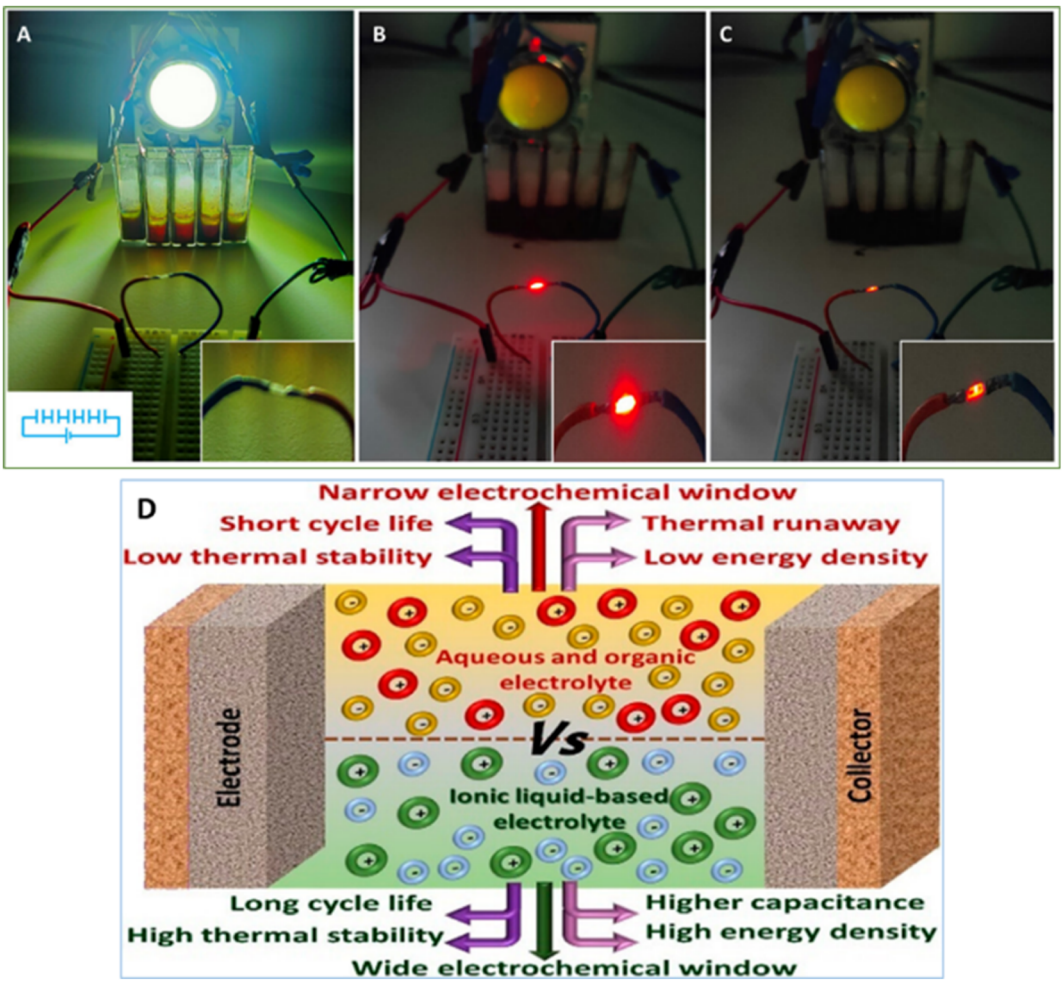


Fig. 6 (A–C) Practical utilization of a photoacid–electrolyte supercapacitor. Reproduced with permission from ref. 82. Copyright 2025, Wiley-VCH. (D) Aqueous and organic electrolytes *versus* ionic liquid-based electrolyte. Reproduced with permission from ref. 88. Copyright 2024, the American Chemical Society.

Furthermore, the development of photoactive electrolytes, such as those based on photoacids, enables direct solar energy harvesting and storage within a single device, as demonstrated by a supercapacitor array that remains powered for an extended period after illumination, highlighting the potential for compact, efficient photo-rechargeable systems.

Bromide ions, in combination with suitable supporting electrolytes, can be used to increase the capacitance of photo-assisted rechargeable zinc-ion hybrid supercapacitors. The most popular aqueous electrolytes are favored for their high ionic conductivity and low cost. Ionic-liquid-based electrolytes are particularly attractive for high-temperature energy storage applications because of their low volatility, high thermal stability, non-flammability, and low-pressure vaporization.[85] Plate-like electric double-layer capacitors (EDLCs) store charge through the adsorption of ions at electrode interfaces, rather than through a redox process, and can thus offer greater cycling stability and cycle life than other energy-storage technologies.

The EDLC equation is shown below:

$$C = \frac{\varepsilon A}{d}$$

Furthermore, the electrolytes used in PSCs exhibit high photochemical stability, preventing degradation under continuous light exposure. Additionally, illumination-induced photochemical reactions in electrolyte species potentially result in unwanted side reactions or decomposition products that minimize long-term stability and device efficiency.[90] Hence, gel polymer electrolytes and stable aqueous electrolytes (*e.g.*, Na_2SO_4 and KOH) are often preferred because of their resistance to photodegradation.[91]

In addition, optical transparency of the electrolyte is important in PSC devices because parasitic light absorption by the electrolyte can reduce the amount of light reaching the photoactive electrode.[92] Moreover, highly transparent electrolytes reduce photon loss and facilitate efficient excitation of the semiconductor photoelectrode, thus improving overall photocharging efficiency.[93] Additionally, electrolytes containing redox mediators (*e.g.*, triiodide/iodide or quinone-based systems) facilitate photogenerated charge transfer between the storage layer and the photoelectrode.[94] Moreover, their photochemical stability must be carefully controlled, as unwanted side reactions or mediator degradation under illumination limit long-term cycling performance. In PSC systems where photothermal effects occur, localized heating under illumination increases diffusion and ionic conductivity within the electrolyte.[95] This accelerates ion transport and minimizes internal resistance, leading to improved charge–discharge kinetics. Similarly, electrolyte pH plays a significant role in semiconductor stability at the electrode–electrolyte interface.[96] For example, materials such as ZnO and Cu_2O can undergo surface degradation or dissolution in strongly alkaline or acidic environments.[97] Hence, selecting electrolytes with appropriate pH and buffering capacity is necessary to maintain a stable photoelectrode performance during long-term operation.

Table 5 Literature matrix of electrolyte systems and charge storage mechanisms in photo-assisted rechargeable supercapacitors

Ref.	Electrolyte/material	System/device type	Key advantages	Charge storage mechanism	Limitations/challenges
74	Aqueous Zn–Br_2 electrolyte	Zn–Br_2 battery systems	High theoretical capacity and discharge voltage	Redox reactions of Zn^{2+}/Br^-	Shuttle effect and slow redox kinetics
77	Binary redox electrolyte species	Hybrid electrochemical storage devices	Improved Zn anode longevity and conductivity	Enhanced electrochemical window	Complex electrolyte management
86	Aqueous electrolyte with carbon electrodes	Electric double-layer capacitors (EDLCs)	Long cycle life and high stability	Ion adsorption at the electrode interface (EDL formation)	Lower energy density compared to batteries
87	Polybromide electrolytes	Zn–Br energy storage systems	Improved ionic conductivity	Bromide redox conversion	Polybromide shuttle effect
50	Bromide ions with supporting aqueous electrolytes	Photo-assisted Zn-ion hybrid supercapacitor	Increased capacitance and improved ionic conductivity	Redox-assisted charge storage	Electrolyte stability issues

Electrolytes need to demonstrate high optical transparency in the visible spectral region to reduce parasitic light absorption and confirm efficient illumination of buried photoactive electrodes.[98] Additionally, quasi-solid gel electrolytes and transparent aqueous electrolytes are particularly advantageous because they enable maximum photon penetration to the photoelectrode surface, improving photocarrier generation and overall photocharging efficiency (Table 5).[99,100]

4. Integration and device engineering

Solar-assisted batteries comparable to or supercapacitors with hybrid traits have also demonstrated high energy densities.[51] Device engineering and integration convert material advances into operational photo-assisted supercapacitor (PSC) infrastructure by optimizing interfaces, fabrication, and architecture; thus, light-driven charge generation can be efficiently translated into stored electrochemical energy. This achievement demands a holistic design of photoelectrode assembly, device geometry, current collectors, electrolyte/interface chemistry, and packaging to reduce electrical and optical losses during enhancement of transport, storage, and charge separation.[101]

4.1 Architectural strategies

PSC architectures can typically be classified into three categories: tandem systems (a traditional supercapacitor charged by a discrete photovoltaic (PV) unit); "all-in-one" or integrated devices (in which the storage layers and light harvesting are unified into a single stack); and asymmetric designs (in which one electrode is photoactive while the counter electrode is solely optimized considering charge storage). All-in-one integrated designs can simplify packaging and reduce interfacial contact losses; however, they require more precise interface engineering and band alignment to prevent carrier loss and rapid recombination.[102] Additionally, all-in-one integrated designs provide compact structures, improved charge transfer, and reduced wiring losses by combining energy storage and light harvesting in a single device.[103] Furthermore, they need band alignment and a precise interface. Apart from that, tandem configurations offer higher individual component efficiency and flexibility but suffer from larger device size, additional energy losses, and increased interconnection resistance.[104]

A bifunctional photo-supercapacitor is illustrated in Fig. 7, in which a supercapacitor and solar cell are unified into a single device. PEDOP@MnO_2 and TiO_2/hb/CDs electrodes are utilized within this architecture to efficiently store and convert solar energy into electrical charge. Under illumination, this configuration delivers a stable voltage output, demonstrating direct solar-to-charge energy conversion (Table 6).

4.2 Photoelectrode engineering

At the same time, the photoelectrode must exhibit robust solar absorption, a favorable energy band position for charge separation, and facile electronic coupling or electrochemical activity with a storage layer. This implies core–shell nanostructuring, 3D architectures (nanosheets, porous scaffolds, and nanowires), and heterojunction formation (*e.g.*, oxide–carbon or oxide–sulfide junctions) that increase the active surface area and enhance light scattering. These nanostructured electrodes enhance photocurrent collection and provide diffusion pathways for ions, thereby improving the photo-enhanced rate capability and capacitance.[106,107]

Fig. 8 demonstrates the energy band alignment and photoelectrochemical mechanism at a semiconductor–electrolyte interface. Under light illumination, electrons are excited to the conduction band, generating electron–hole pairs that drive redox reactions. Furthermore, efficient band alignment fosters charge separation and reduces recombination losses. Moreover, the comparison suggests enhanced photocurrent under illumination, suggesting improved energy storage performance and charge transfer in photo-assisted supercapacitor systems.

Fig. 9 illustrates two primary charge storage mechanisms in electrochemical systems: capacitive and faradaic processes.[110] In capacitive storage, charge accumulation occurs *via* electrostatic adsorption of ions at the electrode–electrolyte interface without involving charge transfer reactions, enabling rapid and

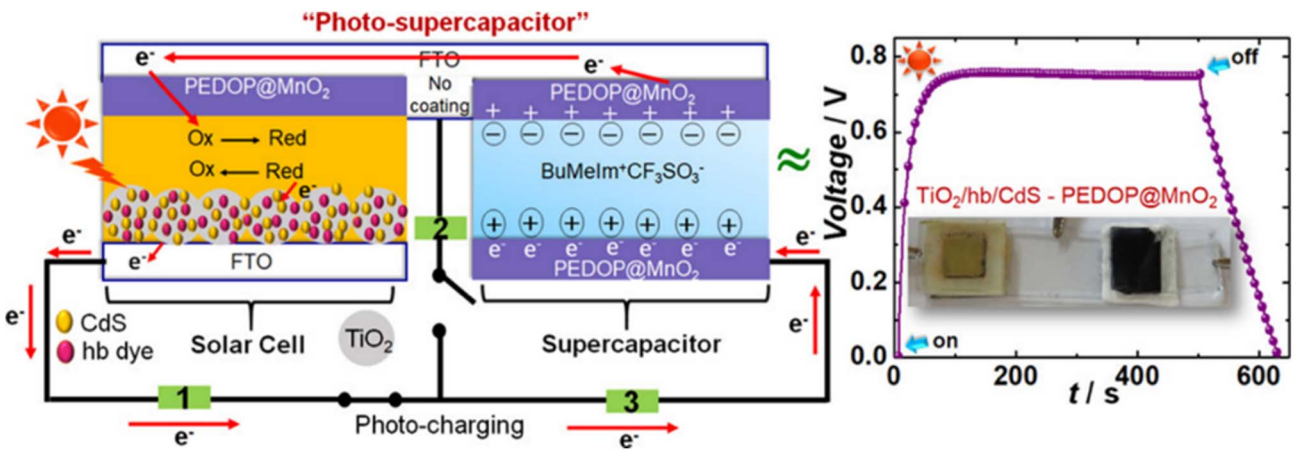


Fig. 7 Illustration of a newly designed bifunctional photo-supercapacitor that integrates solar energy conversion with charge storage. Reproduced with permission from ref. 64. Copyright 2018, the American Chemical Society.

Table 6 Systematic comparison of different photo-assisted supercapacitor (PSC) architectures

Ref.	Architecture type	Photo-charging/ conversion efficiency	Structural description	Energy density	Fabrication complexity	Cycling stability	Key limitations	Key advantages
Flores-Diaz *et al.*[105]	Tandem PSC (PV + supercapacitor)	5.26–11.5% overall solar-to-storage efficiency	Solar cell and supercapacitor connected as separate units through an external circuit	Typically 5–20 Wh kg^{-1}, depending on SC unit	Moderate	>90% after 5000 cycles in many SC systems	Energy losses during energy transfer between modules	Simple design; independent optimisation of PV and SC units
Pandit *et al.*[17]	All-in-one/monolithic PSC	Up to 20.53% solar conversion-storage efficiency	Photovoltaic and storage components integrated into a single device stack	~6.85 μWh cm^{-2} (areal energy density reported for integrated devices)	High	Stable operation over several thousand cycles	Complex interface engineering and band alignment	Reduced internal resistance and compact device architecture
Tuc Altaf *et al.*[51]	Asymmetric photo-supercapacitor	Photo-enhanced capacitance improvements of 30–100% under illumination	One electrode is photoactive, while the counter electrode is optimized for charge storage	Up to ~60.9 Wh kg^{-1} reported for asymmetric PSC systems	Moderate	80–90% capacitance retention after 5000 cycles	High energy density due to pseudocapacitive electrodes	Stability issues and photo-induced degradation are possible

reversible energy storage. In contrast, faradaic storage involves redox reactions at the electrode surface. When ion transport is slower than the reaction rate, the process is diffusion-limited, whereas in pseudocapacitive behavior, fast surface reactions dominate and are not constrained by ion diffusion, resulting in enhanced charge storage kinetics.

Fig. 10 presents an all-solid-state, flexible CoCN//CoCN supercapacitor, highlighting its photo-irradiation enhanced capacitance (PIEC) behavior. Fig. 10(a) illustrates the device concept, while Fig. 10(b) demonstrates the flexible device. The supercapacitor in Fig. 10(c) demonstrates excellent stability, with 83.3% capacitance retention after 5000 cycles. Fig. 10(d) shows cyclic voltammetry data, Fig. 10(e) presents the galvanostatic charge/discharge curves, and Fig. 10(f) shows the electrochemical impedance spectra, confirming the PIEC effect; photo-irradiation (solid lines in Fig. 10(e)) potentially improves the device performance compared to routine operation (dotted lines in (e), blue/red data in Fig. 10(f)).

4.3 Interface and band-alignment control

It is crucial to minimize recombination at the photoactive/electrolyte interface. Engineers regulate band offsets using surface treatments, the introduction of interlayers (ultrathin metal oxides, conductive carbon, or organic linkers), and doping to create favorable built-in potential landscapes that drive carrier separation. Conductive additives, including CNTs, MXenes, or graphene, are typically incorporated into several high-performing designs, with the photoactive phase designed to utilize percolation pathways that facilitate electron flow and buffer volume/mechanical changes during the cycling process (Fig. 11).[111]

A photo-rechargeable supercapacitor demonstrated a 1960% capacitance enhancement using a protonated graphitic carbon nitride and an ambipolar MXene ($Ti_3C_2T_x$) interface, which was further improved under 420 nm illumination. An anomalous capacitance, boosting the photo-assisted voltage to ~270 mV, was facilitated by the interface at ultra-high scan rates in the self-powered supercapacitor.

The spatial variation in binding energy across the device indicates changes in band alignment under dark and illuminated conditions, reflecting differences in the internal electronic structure of the device.[112] The resulting photopotential distribution highlights how charge separation and internal electric fields contribute to voltage generation within the solar cell. Fig. 12 demonstrates key semiconductor photoelectrochemical concepts related to photo-assisted rechargeable supercapacitors. Fig. 12(a) shows the flat-band potential, which indicates the equilibrium energy-level alignment controlling charge transfer.[113] Fig. 12(b) illustrates quasi-Fermi level splitting under illumination, representing photogenerated carrier accumulation that drives photo-induced electrochemical reactions.[114]

In photo-assisted systems, efficient charge injection requires a favorable band alignment between the electrochemical storage layer and the semiconductor.[115] Typically, the semiconductor conduction band must lie ~0.2–0.4 eV above the redox potential to drive electron transfer,[116] while the valence

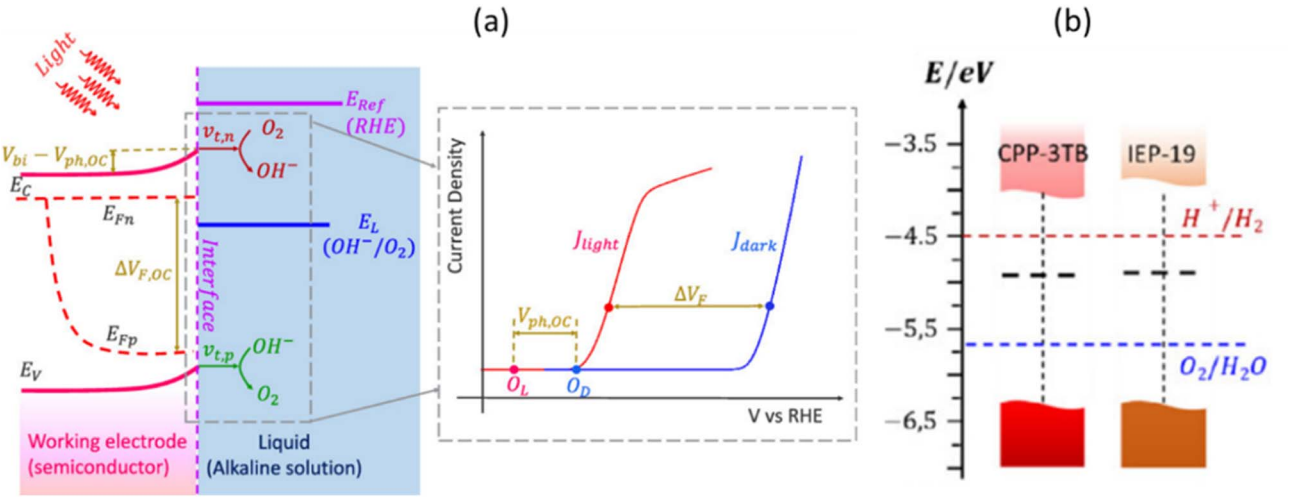


Fig. 8 (a) Energy band diagram and photoelectrochemical mechanism of a photo-assisted electrode (semiconductor–electrolyte interface). Reproduced with permission from ref. 108. Copyright 2019, the American Chemical Society. (b) Energy band diagram illustrating photo-induced charge separation and transfer. Reproduced with permission from ref. 109. Copyright 2025, Wiley-VCH.

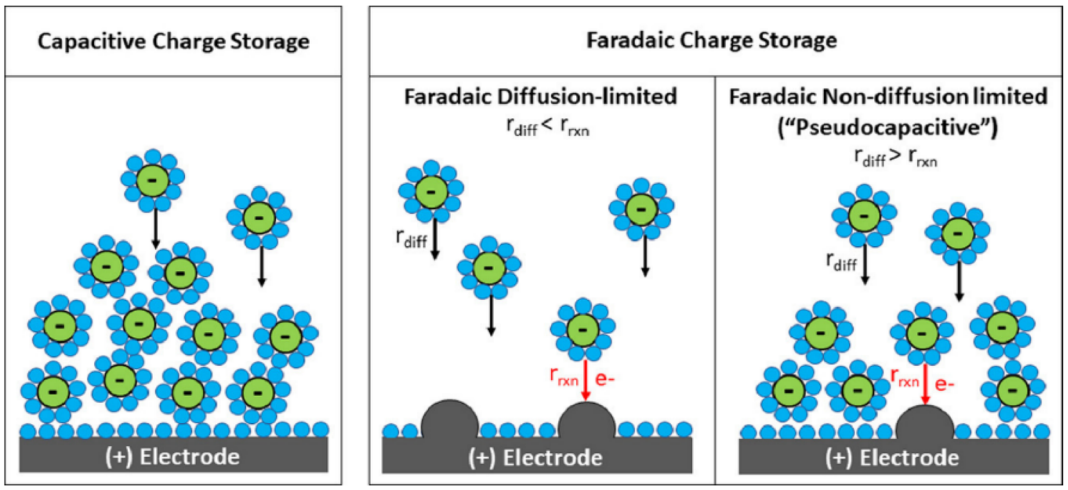


Fig. 9 Schematic of different charge storage mechanisms in electrochemical systems: capacitive charge storage (left), faradaic diffusion-limited processes (middle), and faradaic non-diffusion-limited or pseudocapacitive behavior (right). The figure highlights the relative rates of ion diffusion and redox reactions governing each mechanism at the electrode surface. Reproduced with permission from ref. 110. Copyright 2022, Elsevier.

band must be sufficiently positive to suppress recombination and support oxidation reactions.[117]

Moreover, surface states at semiconductor–electrolyte interfaces remarkably influence charge transfer in photo-assisted devices.[118] Beneficial surface states can facilitate carrier trapping and mediate interfacial redox reactions, thereby improving charge injection. Moreover, excessive defect states act as recombination centers, reducing photocharging efficiency and carrier lifetime.[119,120] Hence, interface engineering and surface passivation are crucial for optimizing the charge-transfer kinetics.

4.4 Electrolyte and redox-mediator design

Electrolytes within PSCs should promote chemical stability, rapid ionic transport under illumination, and compatibility with the redox chemistry of the photoelectrode. Aqueous electrolytes facilitate high ionic conductivity, though they have a restricted voltage window. In contrast, quasi-solid gel electrolytes and ionic liquids have widened the window and enhanced the device lifetime. Redox mediators (*e.g.*, quinines and iodide/triiodide) can commute photogenerated charges and amplify the efficiency of light-to-charge conversion; however, mediator stability during cycling must be engineered to surpass parasitic reactions.[51]

The specific capacitance of the Na-ion supercapacitor can be enhanced through interactions with pre-existing pseudocapacitors. This has been achieved by integrating redox-active electrolyte additives with hierarchical nanostructures of $NaMnPO_4$. The redesign of the conventionally used electrolyte,

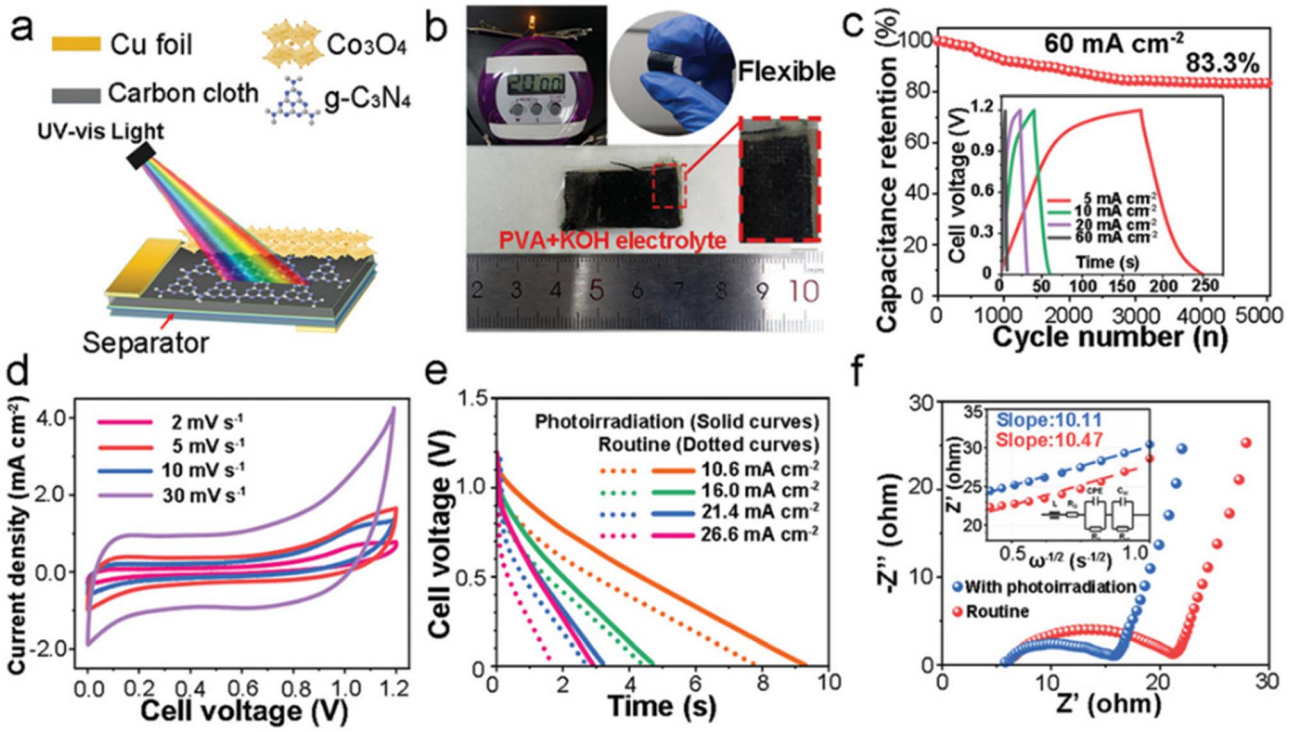


Fig. 10 PIEC performance of all-solid-state CoCN//CoCN supercapacitors. (a) Schematic of the PIEC mechanism. (b) Photographs of a flexible CoCN//CoCN device. (c) Cycling stability evaluated over 5000 charge–discharge cycles; inset shows representative GCD profiles. (d) CV characteristics of the device. (e) Comparison of electrochemical behavior under light illumination and dark conditions. (f) EIS spectra and corresponding fitted curves recorded with and without photoirradiation (inset). Reproduced with permission from ref. 66. Copyright 2020, Wiley-VCH.

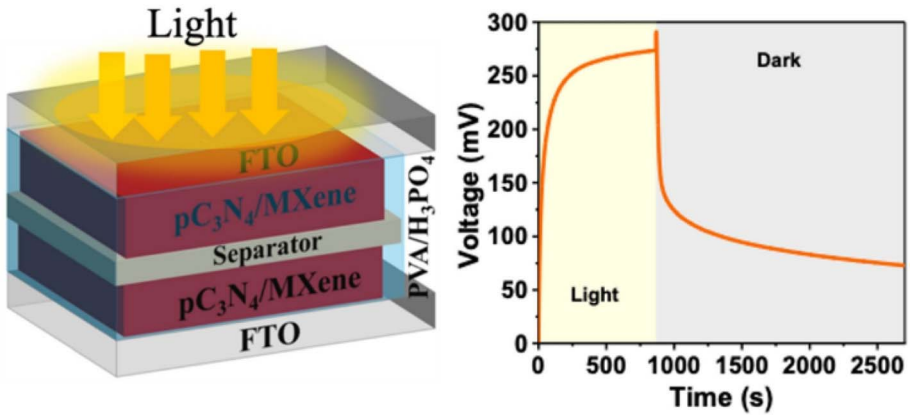


Fig. 11 Self-powered supercapacitor. Reproduced with permission from ref. 111. Copyright 2023, Wiley-VCH.

which involves redox additives (potassium iodide (KI) and potassium ferricyanide [$K_3Fe(CN)_6$]) with NaOH, can achieve an approximately 50% increase in specific capacitance (Fig. 13). It is also possible to retain these improvements in a full cell, fabricated with activated carbon as the negative electrode and $NaMnPO_4$ as the positive electrode. Redox mediators enhance ionic conductivity and facilitate additional redox reactions, thereby improving the electrochemical performance at the electrode–electrolyte interface.

Fig. 14 demonstrates the stability behavior and electrochemical performance of the developed supercapacitor electrode.[82] The curves highlight the charge–discharge characteristics and suggest the improved capacitive response of the material under operational conditions. The results further suggest efficient ion diffusion and rapid electron transfer within the electrode structure, which may contribute to increased energy storage capacity and capacitance. Notably, this stability profile further indicates that the electrode maintains a consistent performance during repeated cycling, indicating good

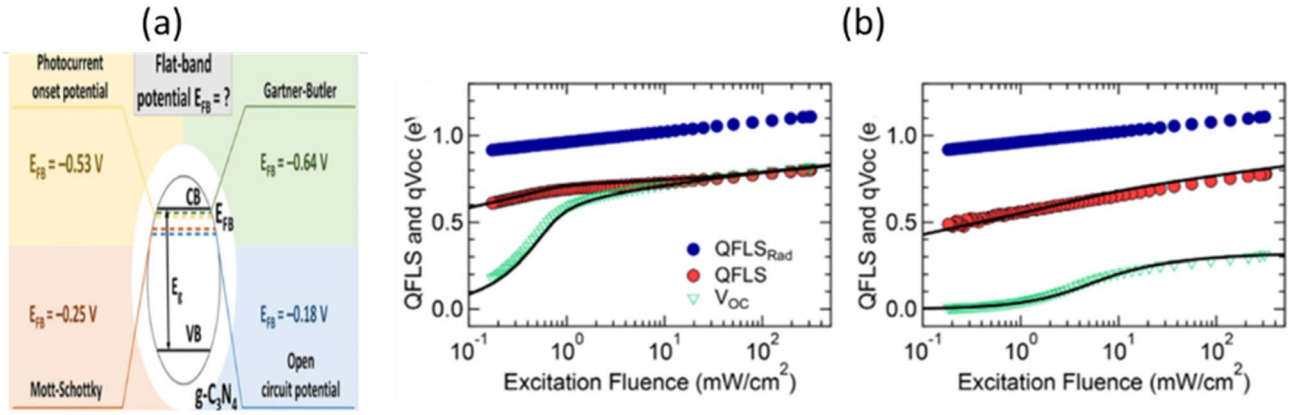


Fig. 12 Band diagrams. (a) Flat-band potential. Reproduced with permission from ref. 113. Copyright 2023, the American Chemical Society. (b) Quasi-Fermi level splitting under illumination. Reproduced with permission from ref. 114. Copyright 2020, Wiley-VCH.

electrochemical reversibility and structural integrity. Finally, the analysis suggests that the material demonstrates promising characteristics for high-performance energy storage applications, especially in photo-assisted or flexible supercapacitor systems where stable conductivity and efficient charge storage are necessary.

Different nanostructured architectures, such as hollow structures, nanotubes, nanosheets, nanowires, nanoparticles, and hierarchical porous frameworks, have been extensively incorporated to enhance the photoelectrochemical and electrochemical performance of photo-assisted rechargeable supercapacitors.[122] Core–shell nanostructures have attracted particular attention because of their unique structural advantages. In these systems, the core typically offers high mechanical stability and electrical conductivity, while the shell provides abundant electroactive sites and improved light-absorption properties. Moreover, this configuration promotes efficient charge separation, enhanced ion diffusion, and rapid electron transport, reducing recombination losses under light illumination. In addition, core–shell architectures effectively integrate semiconductors with conductive materials, improving both

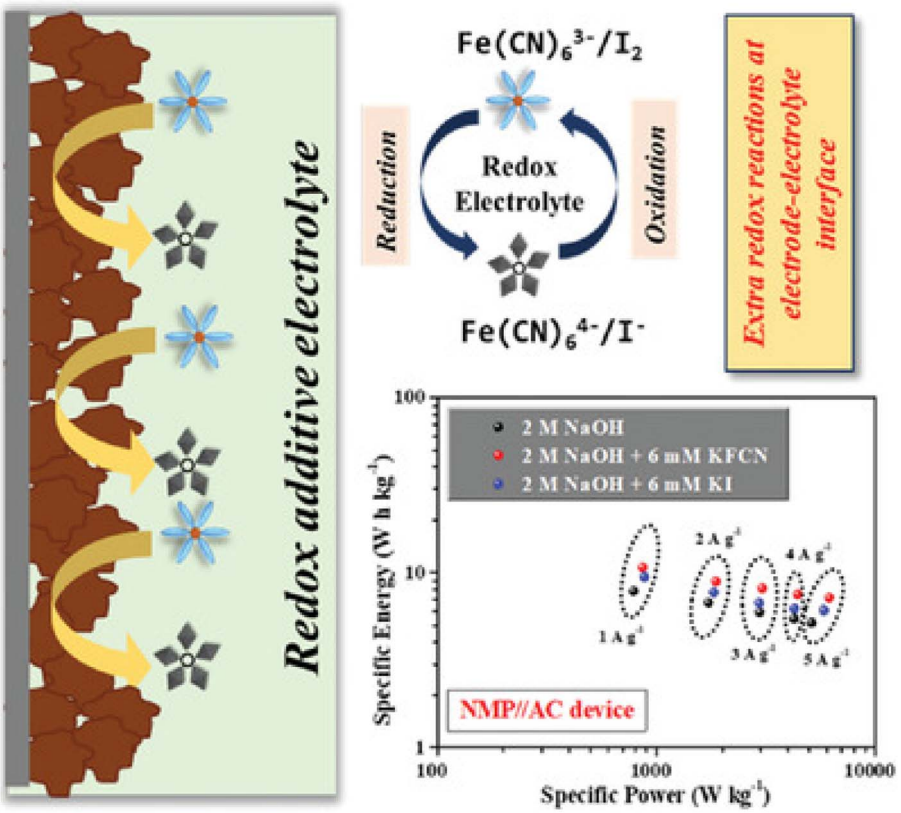


Fig. 13 Redox mediator-induced electrochemical reactions at the electrode–electrolyte interface. Reproduced with permission from ref. 121. Copyright 2021, Wiley-VCH.

electrochemical stability and photocatalytic activity. As a result, the design of advanced core–shell heterostructures is expected to play a significant role in the development of next-generation photo-assisted energy-storage devices and self-powered systems.

The enhancement in electrochemical performance under illumination is primarily governed by the increased availability of photogenerated charge carriers. The reaction rate in photo-assisted systems can be expressed as follows:

$$\text{Reaction rate} \propto n_{\text{photo-generated carriers}}$$

This relationship indicates that higher photon absorption leads to increased electron–hole generation, thereby enhancing the redox reaction kinetics, improving charge transfer, and ultimately increasing the capacitance and energy storage performance of photo-assisted rechargeable supercapacitors.

4.5 Current collectors, transparency, and light management

Transparent conductive substrates (ITO, FTO, and PEDOT : PSS on PET) enable the investigation of buried photoactive layers and are commonly used in integrated PSCs. However, these substrates introduce trade-offs between optical transmittance and sheet resistance. Device engineering has addressed this by using thin metallic grids, ultrathin metal oxide electrodes, or patterned collectors to balance transparency and conductivity. In addition, optical management, including textured surfaces, light-trapping structures, and anti-reflective coatings, enhances photon absorption without compromising the active area.[101]

4.6 Thermal and stability engineering

Continuous operation can accelerate degradation and induce thermal stress (particularly in certain perovskites and organics). Heat-spreading layers, UV filters, the selection of thermally robust photoactive materials, and encapsulation are engineering strategies used to enhance long-term stability. Flexible devices require gel electrolytes and stretchable current collectors to maintain ionic contact during bending cycles.[1]

The concept of photothermal nanomaterials is illustrated in Fig. 15, which demonstrates several nanostructures that convert absorbed light energy ($h\nu$) into localized heat through photothermal effects. These materials, including metallic, semiconductor, and carbon-based nanostructures, play a crucial role

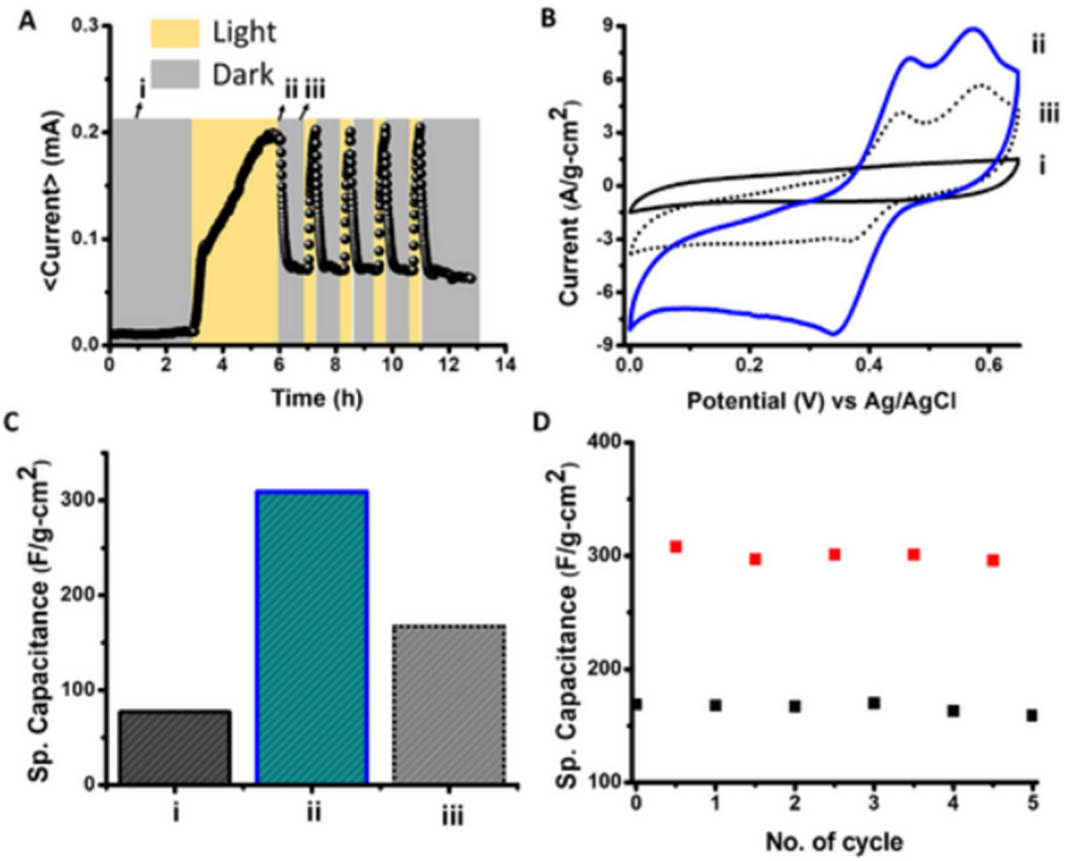


Fig. 14 Light-responsive electrochemical behavior of a 2-nitrobenzaldehyde-based supercapacitor. (A) Transient photocurrent under alternating light/dark conditions showing response before irradiation, during white LED exposure, and after light is switched off. (B) Cyclic voltammetry curves under different illumination states in an acidic electrolyte. (C) Comparison of specific capacitance derived from CV measurements across states. (D) Capacitance stability over repeated cycles in both illuminated and dark conditions, highlighting enhanced performance under light. Reproduced with permission from ref. 82. Copyright 2025, Wiley-VCH.

in enhancing the energy conversion efficiency, thermal management, and light absorption in photo-assisted energy devices.

PSCs degrade during prolonged illumination through several mechanisms. The common degradation pathways include photo-corrosion of semiconductor photoelectrodes, structural instability of electrode materials, photo-induced oxidation, and electrolyte decomposition.[124] Furthermore, continuous light exposure accelerates surface defect formation and charge carrier recombination, reducing cycling stability and photo-charging efficiency.[125] In addition, photothermal effects cause mechanical stress and local heating at the electrode electrolyte interface.[126] To mitigate these challenges, effective strategies such as surface passivation, protective coatings, heterostructure engineering, and the use of gel electrolytes and stable electrolytes are required. Subsequently, implementing robust nanostructured architectures and carbon-based conductive scaffolds improves structural stability and suppresses photo-induced degradation.

4.7 Manufacturing and scalability

To enable the transition of PSCs from proof-of-concept to scalable fabrication, scalable fabrication methods (screen/inkjet printing, spray coating, electrodeposition, and roll-to-roll deposition) can be adapted to deposit storage and photo-active layers on flexible substrates. Photo-supercapacitors and other photo-powered energy storage devices have great potential for smart electronics due to their ability to simultaneously convert and store solar energy. New trends indicate that they have become more flexible, energy efficient, and easier to fabricate, and can be used in next-generation portable power systems and wearable devices. Ensuring uniformity, reproducible electrochemical and optical, and adhesion characteristics over large areas has become an important engineering problem.[127] Moreover, cost-effective materials and low-temperature processing are especially relevant for flexible and wearable applications.

4.8 Performance optimization and diagnostics

Experimental optimization is coupled with *in situ* diagnostics and modeling (transient photocurrent/photovoltage mapping, *operando* spectroscopies, and photoelectrochemical impedance spectroscopy) in device engineering to identify ionic bottlenecks, degradation pathways, and recombination sites. Quantitative values, such as photo-enhanced capacitance, light-to-stored-energy efficiency, repeated photo-charging cycle life, and coulombic efficiency under light, guide design solutions.[101]

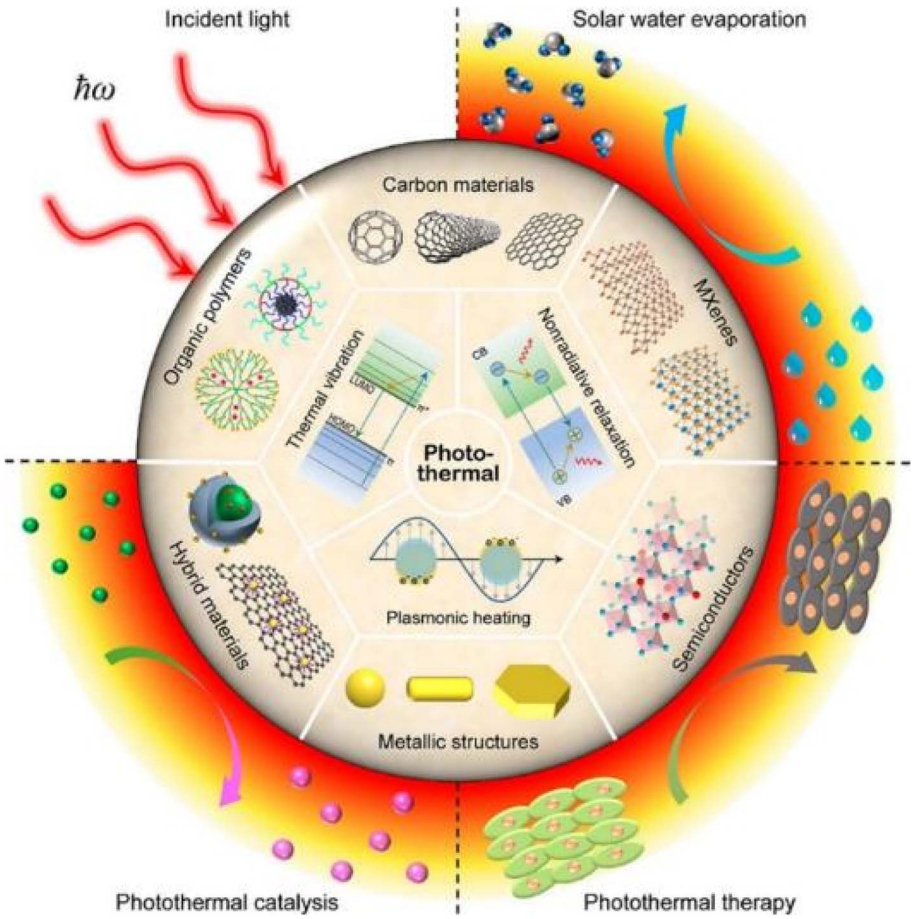


Fig. 15 Photothermal nanomaterials. Reproduced with permission from ref. 123. Copyright 2023, the American Chemical Society.

PSC technologies are still largely at the laboratory scale, and thus precise cost-per-unit energy estimates remain limited. Moreover, preliminary assessments indicate high costs for PSC systems due to complex nanomaterial integration and synthesis steps. In contrast, conventional solar cell-supercapacitor tandem systems typically benefit from mature, lower-cost manufacturing.[128] In PSCs, future cost reductions are expected through scalable fabrication methods, including electrodeposition and printing, and the use of earth-abundant materials, which could potentially improve economic competitiveness for wearable and distributed energy systems.

4.9 Outlook and bottlenecks

Photo-assisted rechargeable supercapacitors require the effective integration of photoresponse, charge-storage density, and cycling stability, which are often at odds with each other; for example, enhanced light absorption can increase ion transport. Stable broadband photo-absorbers, stable heterojunctions with low interfacial recombination, standardized photo-charging measurement systems, and cheap, scalable fabrication methods are therefore key research priorities.[51]

The performance at the nanoscale relies on nanoscale interface engineering, optimized device designs, improved electrolytes, and manufacturable designs to guarantee efficient storage and utilization of photogenerated charge carriers.

Recent advancements in machine learning (ML) and artificial intelligence (AI) have opened new opportunities for accelerating the optimization and discovery of materials for photo-assisted energy storage systems.[129,130] ML algorithms can analyze large datasets of material properties to predict optimal bandgap energies, charge transport characteristics, and electrochemical activity of photoactive materials.[131] Furthermore, these computational approaches allow rapid screening of candidate materials, reducing the cost and time associated with traditional trial-and-error methods. Subsequently, AI-driven modelling can help optimize nanostructure design, electrode–electrolyte interfaces, and heterojunction formation, which are critical for improving charge-separation efficiency and light absorption.[132] ML can also be integrated with experimental techniques to predict cycling stability, capacitance behavior, and energy density under different operating conditions.[133] Thus, AI-assisted materials discovery and device optimization are expected to play a significant role in the future development of high-performance photo-assisted rechargeable supercapacitors and other advanced energy storage technologies (Table 7).

5. Photophysical processes governing photo-assisted rechargeable supercapacitors

The photophysical processes at the photoelectrode–electrolyte interface strongly control the performance of photo-assisted rechargeable supercapacitors. Absorption of light that contains more energy than the semiconductor bandgap produces electron–hole pairs or excitons in the photoactive material.[134] Moreover, the process of exciton dissociation at the interface of the semiconductor should be efficient to generate free charge carriers, which can undergo electrochemical reactions.[135] Also, the heterojunction engineering, band alignment, and surface defects between the photoelectrode and the electrolyte significantly affect the efficiency of this process.[136]

Charging-carrier recombination is another key factor that can reduce the photo-charging efficiency. Recombination is achieved *via* interfacial recombination, surface trap states or bulk recombination at the interface of the semiconductor and the electrolyte.[137] These pathways are frequently measured using time-resolved spectroscopies, such as time-resolved photocurrent measurements, photoluminescence decay, and transient absorption spectroscopy, which provide information on recombination rates and carrier lifetimes.

The connection between the ion transport timescale and carrier lifetime determines whether the device operates in a true photo-charging regime.[138] Notably, when the carrier lifetime created by photo-charging is longer than the hole diffusion time, electrons and the electrolyte can not only drive faradaic reactions but also store energy.[139] Otherwise, illumination primarily increases electrical conductivity, creating a photoconductive effect rather than actual energy storage.

Further, the performance improvement observed under light is due to three processes: photothermal, photoconductive, and photocapacitive processes. Photocapacitive effects involve direct interactions of the photogenerated charges in redox reactions.[140] Photoconductive effects increase the electronic conductivity of the electrodes without retaining additional charge.[141] At the same time, photothermal effects result from light-induced heating, which enhances the conductivity and ion mobility in the electrolyte.[142] Nevertheless, these mechanisms must be distinguished *via* careful analysis using electrochemical impedance spectroscopy, time-resolved spectroscopy, and temperature-controlled measurements.

6. Carrier injection *vs.* recombination timescales in photo-assisted rechargeable supercapacitors

Photo-charging in photo-assisted rechargeable supercapacitors depends on the competition between photogenerated carrier injection into the electrochemical storage layer and carrier recombination. The carrier injection of photoactive semiconductors has been studied using ultrafast spectroscopy and has been found to take place within the femtosecond-picosecond timescales (10^{-15} to 10^{-12} s), whereas recombination reactions occur in the picosecond to nanosecond timescales depending on defect density and material structure. For example, in femtosecond transient absorption studies of doped TiO_2, carrier trapping times of ~1 ps have been observed, which are associated with electron capture states arising from defects.[143]

Additionally, in semiconductor heterojunctions, time-resolved photoluminescence (TRPL) experiments, including those on TiO_2–MXene systems, indicate average photoelectron

Table 7 Literature matrix of integration strategies and device engineering in photo-assisted rechargeable supercapacitors

Author/ref.	Key materials/components	Device/architecture	Key performance contribution	Engineering strategy	Limitations/challenges
84	PEDOP@MnO_2 and TiO_2/hb/CDs electrodes	Bifunctional photo-supercapacitor	Stable voltage output and direct solar energy conversion	Integrated solar-to-charge device	Material stability under long-term illumination
86	CoCN//CoCN electrodes	Flexible all-solid-state PSC	83.3% capacitance retention after 5000 cycles	Photo-irradiation enhanced capacitance (PIEC)	Moderate long-term stability
101	Photoelectrodes, electrolytes, and current collectors	Integrated PSC architecture	Improved charge transport and reduced optical/electrical losses	Optimised device geometry and interfaces	Complex device optimization
107	Core–shell structures, porous scaffolds, nanowires	Nanostructured PSC electrodes	Increased surface area, enhanced photocurrent collection	Photoelectrode engineering	Complex fabrication processes
111	Protonated g-C_3N_4 and MXene ($Ti_3C_2T_x$) interface	Self-powered PSC	~1960% capacitance enhancement under illumination	Interface and band-alignment engineering	Interface recombination and stability
51	PV modules + supercapacitors	Solar-assisted batteries/PSC hybrids	High energy density and improved solar energy utilisation	Hybrid energy storage integration	Energy losses between PV and storage modules

lifetimes of ~3.2 ns, which are remarkably longer than those of pristine TiO_2 (~1.3 ns), suggesting reduced recombination and improved charge separation.[144] Simultaneously, exciton recombination dynamics measured by TRPL in $TiO_2/Ag_2V_4O_{11}$ heterostructures indicated free exciton recombination times of ~1.7 ns and defect-mediated recombination up to ~6.8 ns.[145] These results suggest that true photo-charging occurs when carrier lifetimes (ns to μs) exceed ion-transport times at the electrochemical interface, allowing photogenerated electrons to participate in redox reactions rather than recombining. Therefore, time-resolved techniques, including transient absorption spectroscopy (TAS), are significant for quantifying carrier lifetimes and validating proposed photo-charging mechanisms in PSC systems.[146]

7. Performance analysis

This review summarises the current literature on photo-rechargeable supercapacitors to provide a conceptual and technological overview. Pertinent literature has been gathered from key databases and qualitatively analyzed to highlight key developments in materials, fabrication strategies, and device performance.

Photo-assisted rechargeable supercapacitors (PSCs) exhibit performance parameters that are revealed through the combined assessment of different indices, such as rate capability, specific capacitance (or capacity), light-to-charge storage (or photo-charging) efficiency, coulombic efficiency, long-term cycle stability, and voltage output under illumination. These metrics demonstrate the potential of devices to convert and store solar energy, as well as their functionality under actual operating conditions.

The electrochemical performance of photo-assisted rechargeable supercapacitors is evaluated using key parameters, including specific capacitance, energy density, power density, and coulombic efficiency. These parameters (specific capacitance (C), energy density (E), power density (P), and coulombic efficiency (η)) can be expressed as follows:

$$C = \frac{I \times \Delta t}{\Delta V}$$

$$E = \frac{1}{2} C V^2$$

$$P = \frac{E}{\Delta t}$$

$$\eta = \frac{t_{\text{discharge}}}{t_{\text{charge}}} \times 100$$

where I is the discharge current, Δt is the discharge time, ΔV is the potential window, and V is the operating voltage of the device.

In recent literature, it has been reported that a PSC attached to an ambipolar interface of protonated graphitic carbon nitride

and MXene ($Ti_3C_2T_x$) showed a 1960% increase in capacitance under 420 nm light compared with that under dark conditions, and a photo-assisted voltage of about 270 mV was observed, with capacitive behavior maintained even at ultra-high scan rates.[111] This dramatic improvement shows the potential of suitable photoactive interfaces to amplify charge storage under light.

Research on photo-enhanced magnesium-ion capacitors (photo-MICs) has demonstrated that illumination reduces the charge-transfer resistance (R_{tp}), potentially from ~0.69 Ω in the dark to ~0.20 Ω under illumination, and enhances the capacitance retention at high current densities. For instance, the rate capability increased from ~37.5% in the dark to ~47.2% at 16.2 A g^{-1} under illumination. Cycle stability was maintained under light at ~82.6% over ~400 cycles, demonstrating a stable performance under recurrent dark/light conditions.[147]

An asymmetric PSC comprising dual photoelectrodes ($ZnCo_2O_4$ nanoflowers//hollow $CuCo_2S_4$ spheres) has achieved an energy density of ~60.9 Wh kg^{-1} at 700 W kg^{-1} under illumination, compared with ~46.5 Wh kg^{-1} in the dark. The individual electrode specific capacitances significantly increase under light, as has also been demonstrated in the literature; for instance, the positive photoelectrode achieved 456 F g^{-1} in the dark *vs.* 563 F g^{-1} under illumination (when measured at 1 A g^{-1}).[47] Moreover, the capacitance values reported in the study are expressed as gravimetric capacitance (F g^{-1}), depending on the measurement conditions and electrode architecture.[148]

Cycle-life investigations under illumination are crucial: for instance, resilient self-powered photo-supercapacitors based on PPy–V_2O_5 with tellurium deposition retained ~92–93% capacitance after 12 000 cycles, demonstrating light-induced retention, whereas the capacity retention in the dark is somewhat reduced (but still substantial).[149]

Lastly, the light-to-charge storage efficiency has become a key metric for low-light or indoor conditions. An efficiency of ~21.6% was achieved in a dye-sensitized photo-rechargeable asymmetric supercapacitor using indoor lighting (1000 lux) and MnO–Mn_3O_4/carbon microspheres, the highest reported for an indoor-light PSC.[38]

8. Applications

Solar energy harvesting is seamlessly integrated with PSCs, which offer strong electrochemical energy storage capacity and have a range of potential and emerging applications, especially in self-powered, portable, and off-grid systems.[150,151] A notable field of application is solar-driven asymmetric energy storage, in which usable electrical energy can be directly generated by using dual photoelectrodes. For example, a PSC demonstrated an energy density of approximately 60.9 Wh kg^{-1} under illumination with hollow $CuCo_2S_4$ spheres as the negative electrode and $ZnCo_2O_4$ nanoflowers as the positive electrode, which is higher than its performance in the dark, providing efficient storage in a single device while enabling solar-energy capture.[47]

Fig. 16 presents the sustainable-use ecosystem of PSCs, in which PV units can be used with PSCs to simultaneously generate and store solar energy.[152] Moreover, the circular model emphasizes the interrelationships among system integration, performance, components, and materials, and ultimately enables sustainable applications such as IoT devices, medical devices, wearable electronics, smart homes, and electric vehicles. Combining fast charge–discharge performance with light-driven self-charging, PSCs can offer compact, environmentally friendly solutions to realize off-grid and decentralized energy systems.

Fig. 16 suggests important electrochemical processes, such as photo-reactive redox processes, electrode–electrolyte interactions, and ion transport, which dictate device operation. Furthermore, several upcoming issues in research include scalability, material stability, interfacial charge recombination, and photo-conversion efficiency. Next, the outer ring indicates future work directions, pointing toward optimized material engineering, integrated devices, and improved electrode structures.

Another major use of the Internet of Things (IoT) and wearable electronics is in integrated all-in-one devices. These devices have many advantages, including flexible, photo-reactive, lightweight supercapacitor designs that can collect ambient or indoor light. Most current developments reported in reviews have focused on transparent electrodes, hybrid materials, and defect engineering, which have enabled these photo-charged, flexible storage units to remain continuously operational on top of displays, wireless modules, and miniature power sensors when exposed to light.[51]

PSCs have also been investigated for smart backup power and energy systems, where the grid supply is particularly erratic.[153] Under illumination, their fast charging/discharging makes them appropriate for applications necessitating short bursts of power, including remote environmental sensors, off-grid installations, or emergency lighting.[48] The energy storage capacity from direct sunlight eliminates system complexity and elevates resilience.

Hence, indoor ambient light applications have informed another frontier.[101] Low-light-optimized PSCs could enable capable indoor devices, such as low-power displays, smart home electronics, and wireless sensors, to operate reliably under indoor lighting conditions rather than only when abundant sunlight is available.

9. Standardized performance metrics and testing protocols

To allow meaningful comparisons across PSC research, the research community needs to adopt standardized testing protocols and performance metrics similar to those applied in electrochemical energy storage and photovoltaics.

9.1 Standardized metrics

Researchers need to consistently report photo-enhanced specific capacitance (mF $cm^{-}2$ or F g^{-1}), photo-charging efficiency, coulombic efficiency, power density (W kg^{-1}), and energy density (μWh cm^{-2} or Wh kg^{-1}).[154] Additionally, solar-to-stored-energy conversion efficiency needs to be defined as the

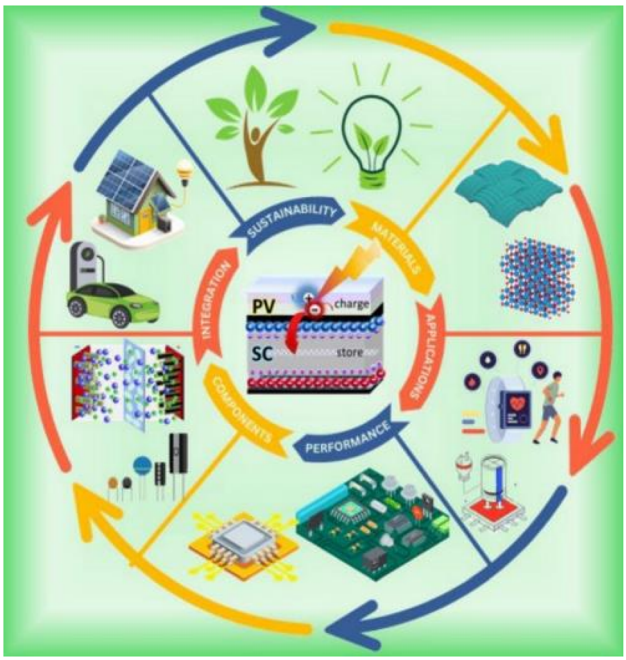


Fig. 16 Sustainable applications of photo-assisted rechargeable supercapacitors. Reproduced with permission from ref. 152. Copyright 2025, Elsevier.

ratio of stored electrochemical energy to incident optical energy.[155] Furthermore, spectral metrics, including IPCE and IQE, should be reported when photoelectrodes are used.

9.2 Standard testing protocols

The measurements should be performed under normal "AM1.5G illumination (100 mW cm^{-2})" to simulate realistic solar conditions.[156] The spectrum, area, and intensity of the light source should be well defined. Also, electrochemical tests must involve controlled illuminated *versus* dark measurements, the same scan rates or current densities, and reported values for electrolyte composition, electrode area, and mass loading.

9.3 Cycling and stability protocols

Similarly, photo-charging and discharging need to be evaluated over ≥5000 cycles under periodic illumination, with simultaneous monitoring of capacitance, efficiency and retention.[10] Adopting these testing conditions and standardized metrics would greatly improve cross-study comparisons, benchmarking, and reproducibility in PSC research.

10. Challenges and future perspectives

Exceptional advances in photo-assisted rechargeable supercapacitors (PSCs) have been made, but several scientific and engineering issues remain that limit their scale-up. The most critical issue is their low photo-charging efficiency. Most widely used photoelectrode materials, such as WO_3 and TiO_2, have large band gaps, which allow light absorption only in the UV region, thereby limiting their total solar utilization efficiency. Narrow-bandgap semiconductors and heterojunctions can enhance visible-light response but may suffer from charge recombination or poor long-term stability.[101]

Another significant challenge is the inefficient electronic and interfacial charge transfer between the photoactive and capacitive components. To achieve efficient electron transport with minimal energy dissipation, accurate band alignment and a robust surface or interface engineering methodology are required.[51] Moreover, the stability of the electrolyte under illumination is problematic because the electrolyte and electrodes are prone to photo-induced side reactions during long-term operation.

On the technological side, the scalability and manufacturability of PSCs remain underdeveloped. Specifically, their fabrication is highly complex, expensive materials are used, and there is a lack of standardized testing protocols, impeding reproducibility and cross-study comparison.[157] This is partly due to the lack of standardized performance metrics, such as the photo-charge-to-energy conversion efficiency, which prevents meaningful benchmarking. Stable hybrid interfaces, inexpensive and environmentally friendly materials, scalable fabrication methods, and standardized evaluation systems are among the areas of inquiry that future studies should focus on to make PSCs more practical and usable in the real world.

11. Conclusion

Photo-assisted rechargeable supercapacitors (PSCs) represent an emerging trend in electrochemical energy storage, as they integrate light collection and charge storage in a single system. This review has presented the evolution of PSCs from primitive hybrid designs to more sophisticated asymmetric and integrated designs, and how materials, device engineering, and interface design share control over photo-enhanced electrochemical activity. Light-generated charge carriers that become actively engaged in redox reactions and ion transport are added to the system by the introduction of photoresponsive electrodes, thereby altering charge storage behavior. All-in-one, asymmetric, and band-level alignment architectures can reduce recombination loss and increase light-energy efficiency. Additional voltage window expansion and operational stability are achieved through the development of electrolyte chemistry, including redox-active additives and photo-responsive electrolytes. Theoretical modeling coupled with *in situ* characterization has also been valuable for providing insight into photo-induced charge-transfer processes.

Author contributions

Chandu V. V. Muralee Gopi: writing – original draft, writing – review & editing, visualization, investigation, data curation, conceptualization. R. Ramesh: writing – review & editing, supervision, funding.

Conflicts of interest

The authors declare no conflicts of interest.

Data availability

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

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