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CHAPTER 7

Advances in Dye-Sensitized Solar Cell–Integrated Energy Storage Systems: Materials, Architectures, and Applications

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Abstract: Dye-sensitized solar cells (DSSCs) have emerged as promising photovoltaic devices due to their low-cost fabrication, design flexibility, and strong performance under indoor and diffuse light. Recent advancements have expanded their role from stand-alone energy harvesters to integrated photovoltaic–storage systems, combining DSSCs with supercapacitors or rechargeable batteries for compact, self-powered solutions. However, challenges persist, including material degradation under light and thermal stress, electrolyte leakage, and performance mismatches between harvesting and storage units. Research efforts have focused on durable sensitizers, thermally stable redox mediators, and robust encapsulation methods to enhance stability. Innovative approaches such as perovskite-sensitized hybrid

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DSSC–storage devices, plasmonic nanostructures for improved light harvesting, bio-inspired and eco-friendly materials, and flexible/wearable formats address efficiency, durability, and sustainability concerns. Electrolyte engineering with ionic liquids, polymer gels, and quasi-solid systems has reduced volatility without significantly compromising conductivity. Future directions emphasize next-generation copper- and cobalt-based redox shuttles, solid-state integration for monolithic photo-rechargeable devices, and recycling-oriented "design for disassembly." Commercial potential is strongest in indoor and building-integrated applications, where DSSCs outperform silicon in low-light conditions while offering aesthetic customization. Overcoming stability, scalability, and recycling challenges will be key to transitioning from laboratory prototypes to market-ready, long-lasting devices for Internet of Things (IoT), wearables, and smart infrastructure.

Keywords: Dye-sensitized solar cells, Integrated energy storage, Perovskite hybrid DSSC, Plasmonic nanostructures, Sustainable photovoltaics.

1. Introduction

Dye-sensitized solar cells (DSSCs) are an attractive photovoltaic technology owing to their low-temperature fabrication, design versatility, and high efficiency under diffuse or indoor light. Recent progress has shifted their application scope from stand-alone power generation to hybrid energy systems, where DSSCs are coupled with storage units such as supercapacitors or rechargeable batteries [1]. This integration enables simultaneous energy harvesting and storage in a compact form, supporting applications in self-powered electronics, wearable devices, and building-integrated energy solutions [2].

One of the major challenges is **long-term operational stability** under combined light and thermal stress. Organic and metal-complex dyes are susceptible to photobleaching, while dye detachment from mesoporous TiO_2 and electrolyte degradation increase recombination losses [1,2]. Thermal cycling, as per ISOS protocols, can compromise sealing layers, counter-electrode adhesion, and the stability of redox mediators. Volatile solvents like acetonitrile, though effective for ionic transport, tend to evaporate or leak, leading to corrosion and dye deterioration [1-3]. In integrated DSSC–storage systems, mismatches between the photovoltaic maximum-power point and dynamic storage load requirements further complicate performance. Additional limitations include optical shading from current collectors, electrolyte incompatibilities, and accelerated degradation during combined illumination and high-rate cycling [3].

Recent advancements focus on electrolyte engineering with ionic liquids, polymer gels, and quasi-solid electrolytes to minimize volatility while maintaining conductivity [1,2,4]. Copper- and cobalt-based redox shuttles now offer higher potentials and improved thermal resilience. Encapsulation techniques, such as glass-frit sealing and monolithic architectures, have enhanced resistance to prolonged light and heat exposure [4]. Furthermore, advanced integration approaches—wire-connected, tandem, and monolithic photo-supercapacitors or photo-rechargeable batteries—paired with impedance-matching strategies, reduce conversion losses [5].

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Future development will rely on co-optimizing materials and interfaces for durability and efficiency, incorporating robust sensitizers, corrosion-resistant counter electrodes, scalable transparent conductors, and refined stability testing, ensuring DSSCs transition from laboratory prototypes to commercially viable, long-lasting devices [4-6].

2. Challenges and Limitations

2.1. Material degradation under light and heat

DSSCs face complex, coupled degradations driven by illumination (UV/visible) and thermal stress. Prolonged light exposure can photo bleach metal–organic and purely organic dyes, shift their frontier orbital energies, and reduce electron-injection yield. Heat accelerates desorption of dyes from TiO_2, corrosion at counter electrodes, and electrolyte decomposition; together these effects raise recombination rates and lower Voc and Jsc over time [6, 7]. Recent experimental/ modeling studies show that device parameters (TiO_2 thickness/porosity, dye loading, iodide concentration) strongly modulate time-dependent decay, and propose figures of merit to quantify degradation pathways [7]. Thermal cycling to ~85 °C remains a stringent stability hurdle even for cobalt-mediated systems, though monolithic device designs have passed standard ISOS protocols. Materials strategies—robust dyes, strong anchoring groups, UV filters, encapsulation, and thermally stable redox shuttles—remain essential [8].

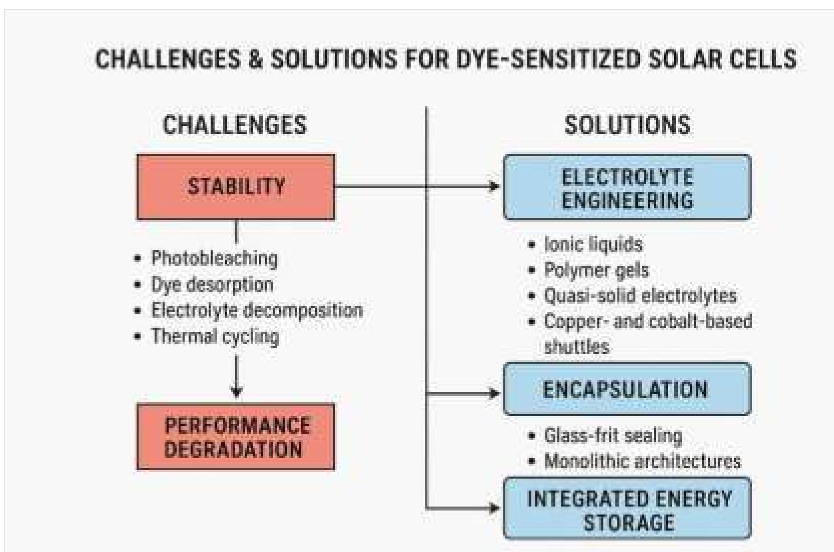

Fig. 1. Challenges and proposed solutions for enhancing stability and scalability of DSSCs.

2.2. Electrolyte leakage and volatility

Liquid electrolytes (iodide/triiodide or Co-based) offer high ionic conductivity but suffer from solvent volatility (e.g., acetonitrile) and capillary leakage through imperfect seals, leading to dye desorption, corrosion, and rapid performance loss [4-7, 9]. Engineering trade-offs are clear: lowering viscosity reduces IR

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drop but can worsen leakage; polymer-gel/aerogel hosts and ionic liquids suppress evaporation and improve mechanical integrity at the cost of slower mass transport. Recent work reports aerogel-based containment and ionic-liquid formulations that enhance stability without catastrophic conductivity penalties; cobalt-mediated monoliths also show improved thermal durability [7,8]. Designing redox shuttles with wider potential windows and low corrosivity, combined with robust seals and quasi-solid electrolytes, is the prevailing path [9].

2.3. Scaling issues for integrated storage (DSSC + supercapacitor/battery)

Photo-rechargeable architectures (photocapacitors, PRBs, and all-in-one SCSDs) promise on-board storage but raise scaling barriers: (i) Impedance matching—the PV side operates best near MPP, while storage prefers variable current/voltage profiles; mismatches cause efficiency losses [7-9]; (ii) Area utilization—co-fabrication reduces active PV area or introduces optical shading by current collectors [9,10]; (iii) Electrolyte and interface compatibility—redox shuttles that are optimal for DSSC may degrade SC electrodes/electrolytes and vice versa [10]; (iv) Cycle and calendar life—integrated stacks experience compounded stress (illumination + high-rate cycling), challenging seals and separators [11]; and (v) Manufacturing integration—printing/roll-to-roll routes must align curing temperatures, solvents, and adhesion across dissimilar layers [10,11]. Recent reviews of photo-supercapacitors and integrated solar-cell/supercapacitor devices outline architectures (tandem, wire-connected, monolithic) and emphasize the need for low-loss power management or built-in rectification to scale beyond lab cells [11-12].

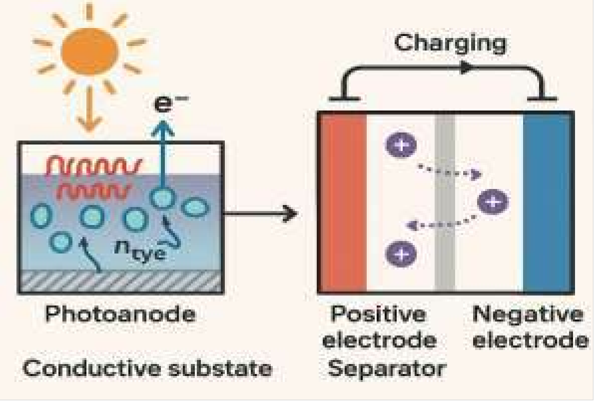

Fig. 2. Schematic of Integrated DSSC–Energy Storage System

2.4. Cost–performance trade-offs

While DSSCs use abundant oxides (e.g., TiO_2, Cu_2O), several high-performance elements drive cost: Pt counter electrodes, ruthenium dyes, hermetic sealing, and precision-coated electrolytes. Replacing Pt with carbon, MoS_2, WS_2, or graphene brings major cost relief but can reduce catalytic activity and long-term stability unless engineered. Similarly, eco-friendly electrolytes and natural/metal-free dyes improve sustainability but often trade off efficiency or durability. Techno-economic analyses increasingly highlight

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materials substitutions and scalable deposition (doctor-blade, screen/inkjet printing) as levers [13]; however, the overall LCOE advantage depends on durability under real-world thermal/UV cycling [13-14]. The literature underscores that pushing to low-cost CEs and benign electrolytes is viable, but reproducible >10-year lifetimes with high PCE and integrated storage remain the decisive milestones for commercialization [14].

3. Recent Advances storage in DSSCs

Integrated dye-sensitized solar cell (DSSC)–energy storage systems represent a promising approach to delivering self-powered, compact, and efficient devices for portable and wearable applications. By combining light harvesting and on-site charge storage within a single architecture, these systems eliminate the need for separate storage units and reduce energy losses during transfer. Recent advances have focused on enhancing performance through diverse strategies: incorporating perovskite sensitizers in hybrid DSSC–storage devices to boost efficiency, employing plasmonic nanostructures to improve light harvesting, using bio-inspired and eco-friendly materials to promote sustainability, and developing flexible, wearable formats for real-world adaptability [15]. These innovations address critical challenges such as power density, durability, and environmental impact, moving DSSC-storage technology closer to practical implementation. Ongoing research emphasizes improving stability, optimizing material interfaces, and scaling up manufacturing to meet the demands of next-generation electronics, particularly in the context of the Internet of Things and sustainable energy solutions [16].

3.1. Integrated perovskite-sensitized hybrid DSSC–storage approaches

Hybrid architectures that incorporate perovskite absorbers with dye-sensitized device concepts or directly couple perovskite photoabsorbers to on-chip storage elements (supercapacitors or thin-film batteries) have emerged as a high-impact direction [15-16]. These systems exploit the outstanding light absorption and high open-circuit voltages typical of perovskites while retaining the mechanical and electrolyte design flexibility of DSSC-style packaging. Practical implementations route photogenerated electrons through low-resistance interlayers into adjacent carbon-based or metal-oxide electrodes that act as supercapacitor charge collectors, enabling rapid photocharging without external power electronics [16,17]. Key engineering challenges that have been addressed recently include interfacial passivation to suppress recombination at the perovskite/electrode boundary, solid-state electrolytes or gel electrolytes to improve stability, and encapsulation techniques that protect humidity-sensitive perovskite components while preserving ion transport to the storage element. These integrated designs show promise for compact, self-charging modules suitable for portable electronics and local energy buffering [15-17].

3.2. Plasmonic nanostructures for enhanced light harvesting and faster charging

Plasmonic strategies are being used to raise the instantaneous power available from photoactive layers so integrated storage elements can be charged more efficiently under weak or diffuse illumination [18]. Carefully engineered metallic nanostructures (gold, silver, aluminum or alloys) placed within or adjacent to the dye layer concentrate near-field intensity, scatter light into higher optical pathlengths inside porous

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photoanodes, and—in some designs—generate energetic "hot" carriers that supplement conventional charge injection [17,18]. Recent work has moved beyond simple nanoparticle decoration to patterned gratings, nanoantenna arrays, and dielectric-coated plasmonic inclusions that minimize parasitic absorption while maximizing scattering and near-field enhancement [16-18]. For hybrid DSSC–storage assemblies, this plasmonic gain translates into improved short-circuit current and thus faster charging of integrated supercapacitors during transient illumination. Practical integration now emphasizes thermal stability and chemical compatibility to avoid plasmonic-induced degradation of dyes and electrolytes [19].

3.3. Bio-inspired and eco-friendly materials for greener, modular devices

Sustainability concerns have accelerated the adoption of bio-derived dyes, cellulose and paper-derived substrates, biodegradable binders, and benign (aqueous or low-toxicity) electrolytes in DSSC-storage research. Natural pigments and co-sensitization strategies can lower cost and environmental impact while enabling low-temperature, roll-to-roll processing for large-area modules [18,19]. Bio-inspired micro- and nano-structures—copied from leaves, butterfly wings, or other hierarchical natural surfaces—improve light trapping and impart antireflective or self-cleaning properties that extend outdoor lifetime [15-17,19]. On the storage side, conductive polymers and bio-based hydrogel electrolytes permit flexible, recyclable supercapacitor electrodes that pair well with green DSSC stacks [18,19]. While efficiencies using natural dyes commonly lag synthetic ruthenium or organic dyes, the tradeoff with recyclability and low embodied energy makes these materials attractive for disposable or low-cost deployments and for devices where sustainability is a prime constraint [17-19].

3.4. Flexible and wearable DSSC–storage devices: system integration and durability

Flexible, stretchable, and textile-integrated DSSC–storage units are rapidly moving from concept toward prototype subsystems for wearables and the Internet of Things [19-21]. Advances include polymer-supported photoanodes, carbon-nanotube or graphene current collectors that combine low bending fatigue with high conductivity, and thin hydrogel or polymer electrolytes that act as both ion conductor and mechanical buffer. Integration strategies laminate thin supercapacitor films directly onto the rear of flexible DSSC stacks or weave energy-storage yarns into fabrics so harvested charge can be stored in place [21]. Research now addresses whole-system issues: minimizing interconnect losses, designing charge-management circuits with ultra-low quiescent draw, and validating cycle life under repeated flexing, washing, and human-contact conditions. Mechanical robustness and sweat/ humidity resistance remain the main hurdles, but recent reports show promising endurance improvements through encapsulation and electrode design [20-21].

4. Future Perspectives

4.1. Next-generation redox mediators

The next wave of DSSC electrolytes is moving decisively beyond iodide/triiodide. Copper(I/II) and cobalt(II/III) polypyridyl complexes now dominate research because they offer higher open-circuit voltages, tunable redox potentials, and markedly reduced corrosivity toward metal grids and catalytic

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counter electrodes [22]. Modern Cu(bpy/phen) systems, in particular, deliver fast outer-sphere electron transfer with low reorganization energy; by ligand engineering (steric bulk, perfluorination, heteroatom donors), researchers suppress recombination at the TiO_2/dye/electrolyte interface while maintaining high diffusion coefficients. Ionic-liquid and gel polymer formulations further enhance safety and lifetime by lowering vapor pressure and leakage risk, a prerequisite for module sealing [22-23]. Looking ahead, expect mixed-valence shuttles (e.g., Cu hybrids), ferrocene derivatives with tailored substituents, and “weakly coordinating” anions to push Voc and stability simultaneously. Under indoor/low-light, these copper systems already show outstanding power densities for IoT nodes, and work on standardizing indoor test protocols will help translate lab gains to product claims [23].

4.2. Solid-state storage integration

A major frontier is merging harvesting and storage so that a DSSC either directly charges an onboard capacitor/battery or is architected as a monolithic photo-rechargeable device. Two practical paths are maturing [24]. First, tightly coupled DSSC–supercapacitor hybrids (wired, shared-electrolyte, or fully monolithic) buffer intermittent light and deliver high pulse power for sensors and wireless bursts; here, minimizing series resistance at the PV–SC junction and selecting high-rate carbon or pseudocapacitive electrodes are decisive [22-24]. Second, integrated photo-rechargeable batteries (IPRBs) adopt two-, three-, or four-terminal layouts to decouple light absorption, charge separation, and storage, thereby reducing energy losses from impedance mismatch and over potential [24]. For DSSC-centric IPRBs, shared redox mediators and transparent current collectors enable compact stacks with good coulombic efficiency. The research focus now is on interfacial selectivity (blocking interlayers to prevent back reactions), voltage matching (e.g., series mini-cells charging Zn- or Li-based microbatteries), and encapsulation strategies that keep both the PV and the storage electrolyte healthy during thousands of cycles [25].

4.3. Recycling and sustainability aspects

Sustainability thinking is shifting from “benign chemistry” to “circular design.” On the materials side, iodine-free copper/cobalt shuttles reduce halogen waste; Pt-free counter electrodes (carbon, transition-metal carbides/nitrides, conductive polymers) cut cost and embodied energy; and solvent systems increasingly favor greener alternatives with lower toxicity and easier recovery. From a device perspective, DSSCs have inherent recycling advantages: the mesoporous oxide and glass substrate are robust and separable, and the liquid/gel electrolyte is recoverable with distillation or membrane extraction [24-26]. Recent reviews outline unit-operation flows for dye extraction, electrolyte regeneration, and FTO-glass reclamation, while life-cycle assessments (LCAs) flag “hot spots” such as solvent use, dye synthesis routes, and module lamination [26]. The near-term goal is “design for disassembly”: crimped or reversible seals, modular gaskets, and dyes with cleavage handles for solvent-free stripping. For manufacturers, publishing product LCAs and adopting take-back schemes will be key to regulatory compliance and customer trust in building-integrated and consumer-electronics markets [23-25,27].

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4.4. Commercialization potential

Commercial traction concentrates where DSSCs are uniquely strong: diffuse/indoor light and aesthetic or form-factor-constrained applications. In indoor PV (hundreds to a few thousand lux), dye/mediator stacks optimized for red-rich spectra routinely outperform silicon on a power-per-area basis, powering Bluetooth beacons, environmental sensors, and e-paper displays [27]. What unlocks productization now is stability: copper mediators, hydrophobic anchors, UV-filtered encapsulants, and low-volatility co-solvents collectively raise operational lifetimes and shelf stability. For modules, scalable screen/gravure printing and laser patterning of series-connected cells enable custom shapes, logos, and semi-transparent façades [28]. Remaining hurdles are well-defined: (i) indoor performance standards and nameplate ratings [26-27], (ii) supply chains for specialized ligands/dyes [26], (iii) field-proven >10,000-hour damp-heat and thermal-cycling reliability [26,27], and (iv) clear recycling pathways for consumer returns [23]. If these are met, DSSCs are poised to grow as "energy-autonomous" surfaces and power sources for IoT, wearables, smart packaging, and building interiors, with BIPV niches where color and translucency matter more than peak outdoor efficiency [25-28].

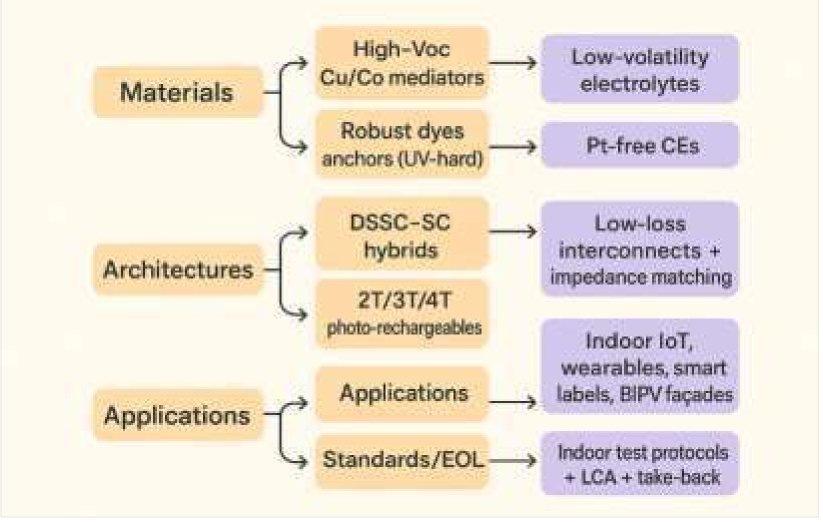

Fig. 3. Roadmap for Future Development of Dye-Sensitized Solar Cells (DSSCs)

5. Conclusion

Dye-sensitized solar cells (DSSCs) continue to advance as a versatile photovoltaic platform, particularly when integrated with on-board energy storage systems such as supercapacitors and rechargeable batteries. Recent innovations in electrolyte engineering, robust sensitizers, plasmonic nanostructures, bio-inspired materials, and flexible architectures have significantly improved efficiency, stability, and adaptability for real-world applications. Perovskite–DSSC hybrids and plasmonic enhancement strategies have pushed light-harvesting limits, while sustainable material choices and recyclable designs align the technology with environmental priorities. Despite these achievements, challenges persist, including thermal and

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photochemical degradation, electrolyte volatility, impedance mismatches in integrated systems, and cost–performance trade-offs. Addressing these requires co-optimization of materials, interfaces, and encapsulation methods, alongside scalable manufacturing routes. Copper- and cobalt-based redox mediators, solid-state or quasi-solid electrolytes, and advanced encapsulation promise longer operational lifetimes under both indoor and outdoor conditions. As research converges on durability, power management, and sustainable production, DSSCs are well-positioned to occupy niche markets where aesthetics, flexibility, and indoor performance outweigh absolute outdoor efficiency. With continued progress toward stable, recyclable, and commercially scalable designs, DSSCs coupled with integrated storage can transition from laboratory concepts to viable solutions for IoT devices, wearables, smart building surfaces, and other applications demanding compact, autonomous energy systems.

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