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Challenges and Perspectives for an Extended Use of the Dye-Sensitized Solar Cells

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ABSTRACT

The upcoming generation of photovoltaic (PV) technologies that will support the required energy shift is expected to include the technology of dye-sensitized solar cells (DSSCs), as DSSCs have been recognized as one of the most promising alternatives to silicon-based devices, ensuring good PV efficiency while exploiting sustainable and low-cost materials. DSSCs are photoelectrochemical cells that represent the first example of a hybrid organic–inorganic photovoltaics system and, as such, initiated the important research sector of BIPV (building-integrated photovoltaics). Recently, DSSCs have also been proposed as an innovative solution in agrivoltaics and archaeophotovoltaics, since the versatility in the chemistry of DSSC components can impart wavelength selectivity in the photoconversion process, a feature that is almost unique in the PV landscape. DSSCs have also demonstrated the capability to efficiently harvest luminous energy under indoor-light conditions, a characteristic that renders them highly attractive for IoT (Internet of Things) applications and, in the context of agrivoltaics, for operation inside greenhouses. Under diffuse light—typical of indoor illumination with artificial light sources—DSSCs exhibit superior photoconversion performance compared to traditional semiconductor-based PV technologies. This advantageous aspect of DSSCs is due primarily to the mesoporous structure of the photosensitive electrode, which enables omnidirectional light harvesting. The present review focuses on the recent evolution of DSSCs, particularly developments in the design, formulation, and preparation of new redox couples, new types of electrolytes, and organic dye-sensitizers. Finally, this contribution also highlights how the nature of the electrode material affects interfacial charge-transfer properties and how electrode surface engineering is crucial in determining both the efficiency and temporal stability of the resulting DSSC. From a production standpoint, recent research has largely focused on identifying scalable manufacturing methods for obtaining DSSCs with high or wavelength-selective transparency for applications in BIPV, agrivoltaics, archaeophotovoltaics, and smart fenestration.

1 | Introduction

Dye-sensitized solar cells (DSSCs) represent a unique class of photovoltaic devices that combine principles of photoelectrochemistry with molecular engineering. First introduced by O'Regan and

Grätzel in 1991 [1], DSSCs as a low-cost alternative to conventional silicon-based solar cells while retaining power conversion efficiencies (PCEs) higher than 10% were further developed especially for applications where flexibility, color tunability, low-light performance (e.g. in indoor or submarine environments,

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vide infra), and transparency (as for building integrated photovoltaics, BIPV, vide infra) are required. Unlike traditional solid-state photovoltaics, DSSCs utilize a molecular photosensitizer to absorb sunlight in the visible range (therefore generally known as dye) and initiate electron injection into a mesoporous n-type semiconductor, typically TiO_2 . The photoinjected electron then percolates through the semiconductor and reaches the anode (acting as a charge collector), while the reduced form of a redox couple regenerates the dye left in the oxidized state after electron injection. The oxidized form of the redox couple, which has been produced at the photoanode, diffuses away and reaches the cathode (counter electrode [CE]), where it is reduced, thus closing the electronic circuit [2, 3].

Over the past three decades, DSSCs have been the subject of intense academic and industrial research due to their potential for low-cost fabrication, use of earth-abundant materials, and compatibility with roll-to-roll processing [4]. The device architecture is relatively simple and modular, consisting of a transparent conductive substrate, a mesoporous semiconductor film sensitized with dye molecules, an electrolyte containing a redox couple, and a CE. This modularity enables precise tuning of individual components and opens up the possibility of application-specific optimization, for instance, BIPV [5], indoor energy harvesting for the Internet of Things (IoT) [6, 7], and semitransparent agri-voltaic systems [8].

Despite significant progress in material development and device engineering, DSSCs have not yet achieved widespread commercial adoption [9]. The record for PCEs of state-of-the-art DSSCs—approaching 15% under simulated sunlight [10] and exceeding 30% under indoor illumination [11]—is certainly encouraging, but several critical barriers to scalability, durability, and cost-effective manufacturing remain. These barriers are closely linked to the challenges associated with all components of the DSSC device, like the redox electrolyte, the dye sensitizer, the semiconductor photoanode, and the CE.

Each of these components must meet a delicate balance of performance criteria. For example, the redox electrolyte must ensure fast dye regeneration and efficient transport toward the CE while remaining chemically stable and warranting an optimal performance, as pursued by recent studies [12–22], including recent works on quasi-solid biopolymer-based systems. In turn, the dye-sensitizer must absorb in a spectral interval of the solar spectrum as broad as possible to maximize the photoinjection of electrons into the semiconductor and maintain long-term photostability at the same time [23–26]. The photoanode must possess a high surface area, warrant the electronic charge percolation route with an efficacious electrical contact at the semiconductor/anode interface. Moreover, the photoelectrode must display the appropriate energy alignment with the dye frontier levels and the redox level as well as minimal charge recombination [27, 28]. The CE must have electrocatalytic properties toward the redox reaction and be chemically compatible with the electrolyte [29–35]. Beside issues correlated with the cell components previously mentioned, also DSSC assembly and encapsulation are not trivial: an opportune strategy of assembly and encapsulation, possibly in a monolithic configuration, should be adopted to prevent cell and PV module degradation due to the exposure to moisture, oxygen, and temperature fluctuations (particularly

crucial factors for outdoor and long-term use), with consequent achievement of long-term stability [36–41].

The recent developments in DSSCs indicate how the field of application of hybrid organic–inorganic photovoltaics is shifting toward applications such as transparent solar windows, wearable electronics, and self-powered sensors, which involve new functional requirements, for example, esthetic tunability, mechanical flexibility, and environmental benignity consequently introduce further complexity. For example, transparent DSSCs require careful modulation of the amount of dye absorbed on the surface (dye loading) and of the redox electrolyte optical properties [42–44], while indoor DSSCs must operate efficiently under low-intensity artificial light sources [45–49].

This review aims to provide a comprehensive overview of the technological solution to the key challenges hindering the large-scale deployment of DSSCs. We focus on the following critical aspects: (1) the limitations and prospects of redox electrolytes, (2) the design and stability of dye sensitizers, (3) issues in the engineering of photoanodes and CEs, (4) the overarching problem of long-term device durability, scalability, and reproducibility, and finally, (5) the design of different types of DSSCs based on less-explored operation principles. By highlighting current bottlenecks and evaluating emerging strategies, we intend to delineate future directions in the development of DSSC technologies for real-world applications.

A few acronyms used throughout the text are briefly outlined here for clarity. PCE refers to the overall capability of the device to convert incident solar energy into electrical output. The incident photon-to-current efficiency (IPCE), also known as EQE, describes the wavelength-dependent response of the cell, indicating how effectively absorbed photons are transformed into electrons. The average visible transmittance (AVT) and visible light transmittance (VLT) quantify the transparency of semitransparent devices within the visible range, which is particularly relevant for building-integrated or aesthetic applications. The light utilization efficiency (LUE) expresses the trade-off between optical transparency and photovoltaic performance, while the color rendering index (CRI) assesses the visual quality of transmitted light in comparison with natural illumination.

2 | Evolution of the DSSCs

In their first appearance, DSSCs were based on a nanostructured photoactive anode [an n-type semiconductor (SC) sensitized with an organic or organometallic dye] and a metallic cathode with electrocatalytic properties toward the electroreduction of the oxidized form of the redox couple, with the electrodes that are contacted by a liquid electrolyte containing the redox couple. The dye-sensitization of wide bandgap SCs having an energy bandgap larger than >3 eV (or, equivalently, absorption wavelengths lower than 420 nm) for the production of photocurrents at luminous energies lower than the characteristic one of unsensitized bulk SC, was first demonstrated by Gerischer in 1968 for both n- and p-type SCs when combined with sensitizers having frontier levels matching with the band edges of the bulk SC [50]. An important step forward on the basis of the successive fast progress

in the fields of semiconductor sensitization and hybrid photovoltaics was the invention of the DSSC by Michael Grätzel and Brian O'Regan in 1991 [1]. The DSSC represents the photoelectrochemical device that started the development of low-cost photovoltaics, thanks to the ability to reach PCE values as high as 15% [3]. The flourishing development of the DSSCs was made possible by the ability to produce nanostructured inorganic SCs as photoelectrodes that are based on chemically stable materials like the oxides of transition metals [51]. Such electrochromic nanostructures of n-type could be sensitized with chemically anchored dyes [52] that, upon visible irradiation, photoinject electronic charge in the nanostructured n-type SC with production of high photocurrent densities (at least 10^3 times larger with respect to a nonnanostructured photoelectrode).

Recent innovations in device architecture—including the use of sensitized nanostructured p-type SCs [53] as photocathodes, composite materials for any of the components of the DSSC, counter-electrodes with improved electrocatalytic properties, new redox couples with an appropriate position of the redox level with respect to the dye frontier levels, faster kinetics of electron transfer with respect to the benchmark I_3^-/I^- couple, and the use of gels or solid electrolytes in the DSSC—have enhanced semitransparency, interface control, wave-selective absorption, and long-term stability, among other aspects. The case of DSSCs with tandem configurations [53–62], in which both nanostructured SC anode and SC cathode are dye-sensitized, constitutes an example of optimization of the PV performance since the presence of two photojunctions allowing simultaneous photoinjection of electrons and holes extends the external quantum efficiency (EQE) on a wider spectral interval with respect to the DSSC with single photojunction with consequent beneficial effect on PCE of the solar radiation. This is the result of a careful alignment of the HOMO-LUMO levels of the sensitizers with the band edges of both SCs and, more importantly, of the complementary absorption features of the sensitizers at the anode and cathode, which generate a synergic effect of sensitization [56, 57, 59]. In fact, the strategic selection of electrode materials and sensitizers in tandem DSSCs leads to gains in performance over single-photoelectrode DSSCs [63]. For instance, the nanocomposite NiO/ZrO_2 cathodes combined with nanostructured TiO_2 anodes have outperformed their individual counterparts [55]. These electrochromic nanocomposites enabled semitransparent tandem cells with the attainment of a favorable current matching and imparted bifacial features of photoconversion [7, 64].

Available scalable methods of deposition can produce nanostructured thin-film electrodes with controlled composition, thickness, porosity, crystallinity, and adhesion properties, which determine the overall DSSCs' optical and photoelectrical behavior [65–68]. Such an availability of scalable fabrication methods for nanostructured electrodes is essential for the implementation of the DSSCs in the field of BIPV and agrivoltaics, both requiring the application of photoconversion devices on very large areas like walls of buildings or agricultural fields. In this perspective, it is envisaged the introduction and the diffusion of photovoltaic systems with spectral selectivity like DSSCs, also in archaeological ambit (archaeophotovoltaics) with the installation of energy supply infrastructures to be collocated in areas of historical/archaeological interest, which are subject to aesthetic, architectural, and historic-contextual

constraints. The archaeophotovoltaics-generated power would improve the fruition of the archaeological areas, with the electrical energy destined to the maintenance of services utilizable in loco. It is evident that the archaeophotovoltaic systems have to pursue the minimization of the visual impact that the installation of a photovoltaic plant unavoidably brings about and, in this regard, spectrally selective, quasi-transparent DSSCs can constitute one of the most appealing solutions in archaeophotovoltaics given the historic preservation orders and cultural heritage constraints in force in archaeological areas.

Techniques like RDS in plasma atmosphere could yield high-quality electrodes [68, 69] at a relatively low energetic cost over traditional deposition methods through a peculiar mechanism of heat flow that exploits the absorption properties in the microwave regime of the materials to deposit. The adoption of RDS led to the improvement of electrochromic properties such as porosity, homogeneity of chemical composition and morphology throughout the film, surface uniformity, grain boundary structure, and electrical connectivity at the interfaces and between nanostructures [70, 71]. Besides the scalability of the methods for electrode production, the implementation of the DSSCs in BIPV and agrivoltaics also requires the scalability of the production methods of the molecularly engineered sensitizers. Since effective sensitizers must feature high extinction coefficients in the solar spectrum, appropriate dynamics of electron exchange with the nanostructured semiconductor and with the redox couple, suppressed recombination of the photoinjected charge, photochemical stability, and chemical anchoring robustness [72, 73], their structure exhibits high complexity. It is here recalled that a sensitizer for DSSCs has to accomplish several tasks, the realization of which implies a multifunctional role for this component. The synthesis of efficacious sensitizers generally requires articulated multistep synthetic procedures with low processability. The latter aspect is crucial for the scalability of the synthesis of the sensitizers. To overcome such a difficulty, a common synthetic strategy is to adopt a converging approach, where building blocks (designed to accomplish a specific function) are independently synthesized and successively assembled via site-specific coupling reactions [74, 75]. The multifunctional sensitizer shown in Figure 1 has been synthesized for a p-type DSSC exploiting such multiblock approach.

The structure of Figure 1 [74] makes it possible to distinguish three key units in the dye structure: the anchoring moiety chemically connecting the dye with the nanostructured electrode, the light absorption and charge transport unit that, upon photoexcitation, allows the electronic charge transfer from an electron-rich to an electron-poor region of the molecule, and, finally, the external unit where the exchange of electrons between dye and redox couple occurs. The π -conjugated structure responsible for light absorption is the most frequently engineered part of the sensitizer since it controls the HOMO–LUMO gap and dye dipole moment change upon light absorption with its extension and nature/position of substituting groups [76]. Anchoring groups—carboxylic, cyanoacrylic, phosphonic acids, and many more—must ensure dye immobilization and efficient interfacial charge transfer [52, 77] through the saturation of the SC surface defects. The stability of the anchoring group and that of its chemical bond with the SC surface strongly affect the activity of the photoactive material of the PV device over its lifetime.

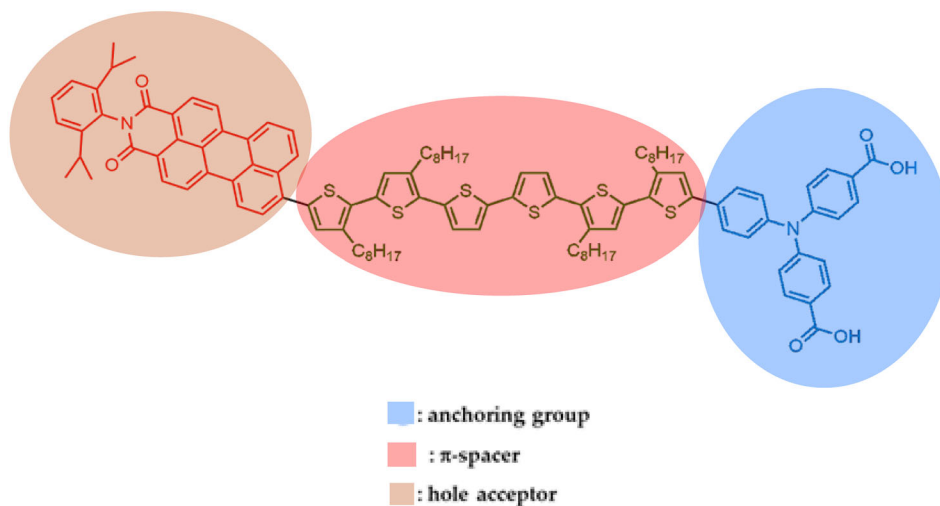


FIGURE 1 | Example of a multifunctional sensitizer with a tri-block structure. The dye features 1) an anchoring group, in this case the carboxylic group —COOH, responsible for the chemisorption of the dye-sensitizer onto the nanostructured semiconductor; 2) a π -spacer constituted by a conjugated moiety (hexathiophene ring in the sketch) separating the site of charge injection (dye/semiconductor interface) from the site of charge transfer with the redox species (dye/electrolyte interface) while warranting charge transport between these two sites; and 3) a moiety containing the charge acceptor, i.e., the moiety that is responsible for the charge transfer between the sensitizer and the redox species.

3 | Dyes for DSSCs: Examples of Established Dyes and Advanced Techniques for Selective Absorption

Being the principal light-absorbing material, the sensitizer is crucial in determining the coloration of the resulting DSSC and constitutes the actual photoactive component in the photo-electrochemical conversion device. Compared to organometallic complexes, representing the first explored class of dye-sensitizers for DSSCs, organic dyes have generally higher molar extinction coefficients and, for this reason, allow the use of thinner and more transparent layers of semiconductors to achieve the same extent of optical absorption. At the same time, the ease of altering the structure of an organic compound, thanks to the synthetic versatility of organic chemistry, opened up the development of dyes with a broader spectrum of coloration with respect to the available portfolio of organometallic sensitizers [78].

One of the most commonly used molecular designs for organic dyes in DSSC is the modular D–A– π –A [79, 80] one obtained by assembling an electron-rich moiety, also known as a donor (D), next to an electron-poor one, acting as an acceptor (A) in the photoexcited state. In addition to that, a conjugated π -bridge links the dyad D–A to an anchoring function with the role of binding the dye to the semiconductor substrate. The electronic structure of the D–A– π –A molecule results modified by the addition of the electronically conjugated π -bridge and can absorb visible light through a HOMO–LUMO transition of a different type (π – π^* , n – π^* , and charge transfer) with respect to the simpler D–A motif, at a smaller energy within the visible range. Implementing the D–A– π –A molecular design with synthetic tools like palladium-catalyzed cross-coupling [81] and direct arylation reactions [82] boosted the exploitation of organic materials in the whole optoelectronic field. When exploring different organic dyes, it is worth keeping in mind that their optical properties are affected not only by the donor [83, 84] and internal acceptor moieties [85] but also by the anchoring function [86, 87], which plays a key role in tuning the optical properties and staining kinetics relevant to

eventual cosensitization processes [88]. By carefully tuning the spectroscopic properties of the dye, DSSC entered specific niche applications inaccessible to other PV technologies, like indoor devices [24] where the absorption spectrum of the sensitizer needs to fit the emission spectrum of artificial bulbs, or BIPV [89–91], for which high perceived transparency is desirable to be exploited in aesthetic partitions, window elements, and other transparent parts of the building envelope. In the latter case, by pursuing devices with high visible transparency and neutral color appearance, the sensitizer is not allowed to absorb the visible radiation in the 500–600 nm range (and absorb little in the 400–700 nm range) where the human eye has its maximum sensitivity. The best strategy to preserve good PV efficiency is to exploit the ultraviolet (UV) and early visible regions (<400 nm) with yellowish dyes, as well as the late visible region approaching the near-infrared (NIR) radiation (>700 nm) through the utilization of green–blue squaraines [92–94] (vide infra) and seemingly colorless sensitizers. In this section, dyes are classified depending on the coloration. It is worth stressing that this classification is not trivial due to the nonlinear and nonbiunivocal correlation between the apparent color and the wavelength range of absorption. However, giving priority to dye color has the pragmatic advantage of clarifying the final color of the fabricated device, which is the most critical aspect of interest for aesthetic and architectural purposes in BIPV.

3.1 | UV-Early Visible Region

Despite the low intensity, the UV-early visible region, spanning from 100 to 450 nm, represents a first slice of very powerful solar radiation that can be exploited to generate photocurrents without interfering with transparency in the visible region of the final device.

For this purpose, hydroxycinnamic acid derivatives, such as caffeic acid (CA), ferulic acid (FA), and *p*-coumaric acid (PA)

(Figure 2A), have been demonstrated to harvest UV-A photons (315–400 nm) with resulting transparency in the visible for the corresponding DSSCs [95]. The photovoltaic performance reached 3.62% under UV-A LED illumination (365 nm, 115.22 mW/cm²), with a corresponding VLT of 49.07%–43.72% and color rendering index (CRI) of 90–93.

The combination of a fluorene core as donor motif, with diverse π -conjugation bridges [1,4-benzene (**FPCN**), 2,5-thiophene (**FTCN**), and 2,5-furan (**FFCN**) rings] brought to a small library of dyes with different photovoltaic efficiencies and transparency, demonstrating the important role of each portion of the π -conjugated structure in the dyes (Figure 2B) [96]. Good efficiencies (PCE = 2.77% under solar simulator and 10.36% under UV-A LED illumination) and transparencies higher than conventional DSSCs (VLT up to 50.11%) were obtained with the dye bearing the **FFCN** π -spacer.

The 2-phenylthiophene core is another interesting UV-absorbing structure. Five different organic dyes were synthesized by Chen and coworkers, functionalizing the 2-phenylthiophene scaffold with alkyl chains of different lengths (**4OPC**, **6OPC**, and **8OPC**) or different positions (**6OP6C** and **2C6OPC**), affecting bulkiness (Figure 3) [97]. The PV performances of the devices were measured under AM1.5 G (100 mW cm⁻²) and UVA LED (365 nm, 150 mW cm⁻²) irradiation, with **8OPC** showing the best performances (PCE = 0.89% and 5.94%, respectively), together with the lowest transparency in the series (AVT = 32.38% for 8 μ m-thick TiO₂ film). AVT was shown to be mostly dependent on photoanode thickness, and over the series, an inverse proportionality between AVT and PCE was observed.

An interesting and practical application of DSSC, dependent on the selective transparency of the dye, is the manipulation of the light spectrum entering the greenhouse [98]. For this application,

a concept of transparency similar to AVT is applied, shifting the reference for sensibility from the human eye (400–700 nm) to the photosynthetic systems (450–520 and 650–700 nm) instead. In this context, slightly yellow UV-selective organic dyes find application. Dyes like the diimides were reported by Alnoman et al. [99], and the curcumin dye reported by Albulescu et al. [100], have been proposed for the exploitation of the light transmitted by a greenhouse. The resulting photovoltaic performances, given the particular setup in which they were assessed (or predicted), could not be compared.

3.2 | Toward Green–Blue Sensitizers

Green and blue dyes are of interest for DSSC in BIPV since they do not absorb light in the 500–600 nm range of the solar spectrum, that is, the range where the human eye is most sensitive, with resulting low perceived alteration of the light intensity passing through the device. Two bands of absorption generally characterize both colorations: while both absorb the red range (600–700 nm), blue-looking dyes also absorb in the yellow-orange range (570–620 nm), while green-looking sensitizers also absorb blue wavelengths (400–500 nm). Some natural materials, for example, indigo, anthocyanines, and chlorophylls, provide access to green/blue-colored molecules that possess both anchoring and electrochemical properties, potentially making them function as sensitizers for DSSCs. Many are reported in the literature [101, 102], among which porphyrin-based sensitizers stand out for the number of examples and the optical properties [103]. However, to overcome disadvantages that occur in natural dyes when immobilized onto a SC substrate, for example, poor intrinsic chemical stability, weak binding with the SC, poor efficiency of intramolecular charge transfer for the nonoptimized orientation of the dye with respect to substrate surface (this consists in the loss of internal charge transfer vectoriality consequent to the

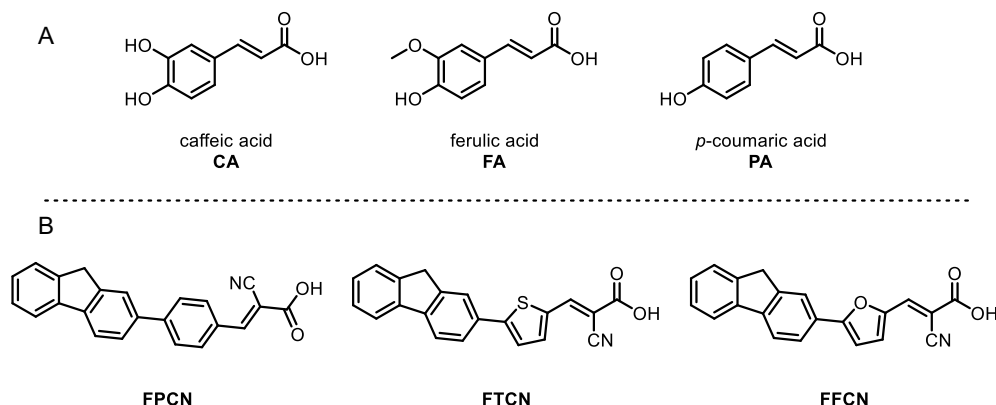


FIGURE 2 | (A) Hydroxycinnamic acid derivatives and (B) fluorene-based cyanoacrylic acid derivatives as sensitizers for DSSCs.

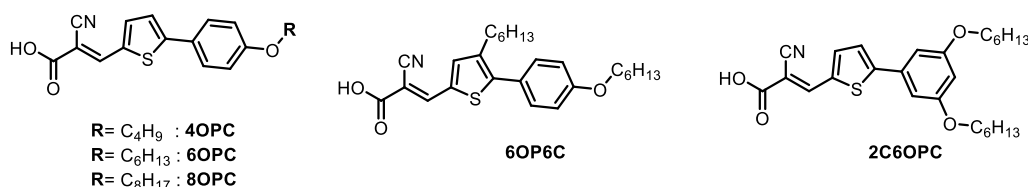


FIGURE 3 | Phenylthiophen-2-yl cyanoacrylic acid-based dyes for semitransparent DSSCs.

existence of strong interactions between the anchored natural dye and substrate), not optimized charge transfer properties between dye and SC/redox mediator for the poor overlap of the dye frontier orbitals with the SC band or the frontier levels of the redox mediator [104–106], several porphyrin-derived dyes can be found that optimize the photovoltaic performances [103, 107], as well as purely synthetic dyes. Among the latter, thieno[3,4-*b*]pyrazine (TPz)-containing dyes have found large applicability for fabricating green-colored transparent DSSCs. The TPz-based symmetric dye **AP3** (Figure 4A) shows an absorption spectrum covering all the visible region with an onset at 800 nm and high transparency in the 500–650 nm range [108]. Such an absorption pattern, together with PCE = 5.0% and an IPCE = 50% peak efficiency, made it a good candidate for BIPV. Such photoconversion performances could be boosted by the cosensitization of TiO₂ SC with a dye having light absorption properties complementary to those of the sensitizer (case of the combination **AP3** sensitizer/**D35** cosensitizer (Figure 4B), as discussed later. A small family of TPz-based dyes was derived later [109]. The characterization of this family of dye-sensitizers for DSSC purposes evidenced that triphenylamine (TPA) was a good donor group, the pyrazine ring was the driver of intense absorption toward the NIR, and the phenyl ring was the best spacer between TPz and the carboxylic acid anchor. Franchi et al. [110] designed a symmetrical D–A–D TPz-based sensitizer (**TP2**) and an unsymmetrical D–A– π –A homologous (**TP1**), proving the need for a symmetrical structure to access high values of transparency (AVT of 42.3% and 83.7% for **TP1** and **TP2**, respectively) with similar PV efficiency (5.16% and 4.03%, respectively)

(Figure 4C). A similar asymmetric dye-sensitizer design that combined TPz acceptor with TPA donor and 2-cyano-3-(2-thienyl) acrylic acid as an anchoring group with electron-withdrawing properties was considered by Lu et al. to develop quasi-solid-state devices [111]. These DSSCs presented stable efficiencies (5.30% for 1000h) but with no assessment of transparency.

Squaraine is another widely exploited central scaffold for the synthesis of dyes, which allows for the exploitation of both blue and green colorations upon small changes in the structure [112, 113]. Examples relevant in the field of DSSC are **SQ-138** and **SQ-140** reported by Roy et al. [114], both reaching AVT values higher than 70% on a 4 μ m thick TiO₂ layer, harvesting photons mainly above 600 nm. The systems **SQ-222** and **SQ-223** reported by Pradhan et al. [115] also rely on a 400–500 nm absorption component, but unfortunately, the corresponding transparency was not evaluated (Figure 5A). Interestingly, in the case of the green dye **SQ-140**, the bifacial configuration was also explored [116], improving the PCE from 3.74% to a cumulative efficiency of 7.17% using an 8 μ m thick photoanode.

Other dyes with similar optical properties have been reported for DSSCs displaying NIR absorption and AVT values worth of attention. The examples of green sensitizers are relatively rare and are mostly obtained from the aforementioned scaffolds, the dithienophenazine [117, 118] and indeno cores [119]. Many more examples are instead reported for NIR-absorbing blue sensitizers that can be obtained from indigo core [120], aza-Pechmann [121] molecular structures, the diketopyrrolopyrrole scaffold (exploited

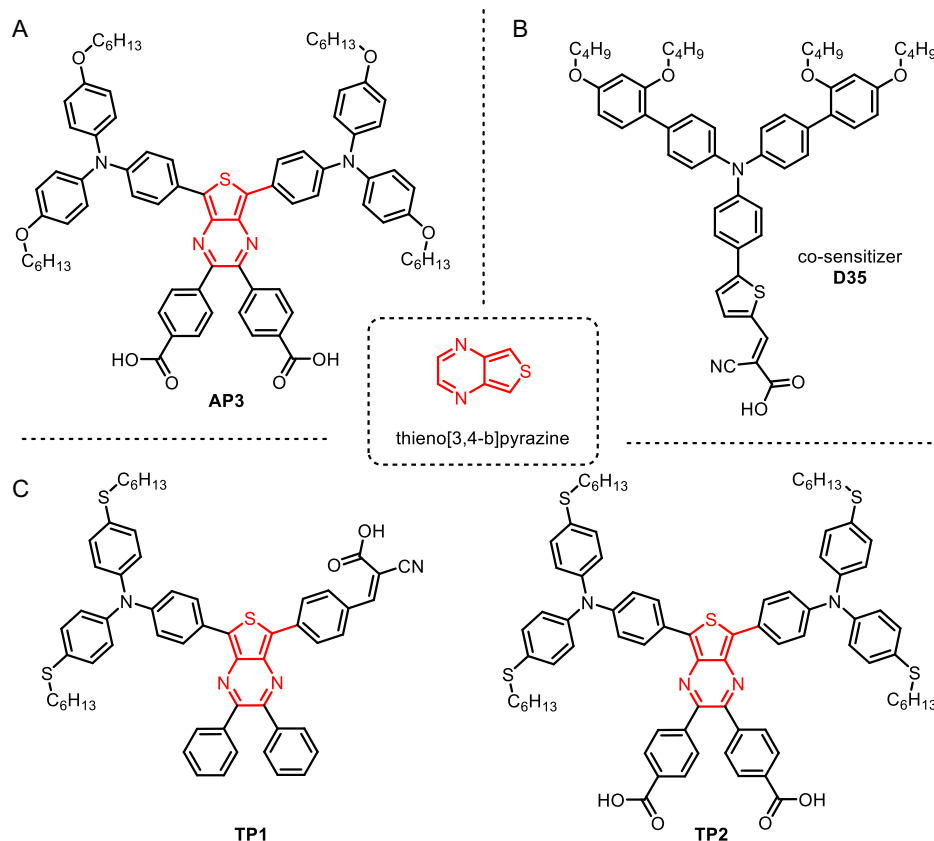


FIGURE 4 | (A) the thieno[3,4-*b*]pyrazine based fluorophore **AP3** and (B) its cosensitizer **D35**; (C) symmetrical (**TP1**) and asymmetrical (**TP2**) thieno[3,4-*b*]pyrazine based fluorophores for panchromatic DSSCs.

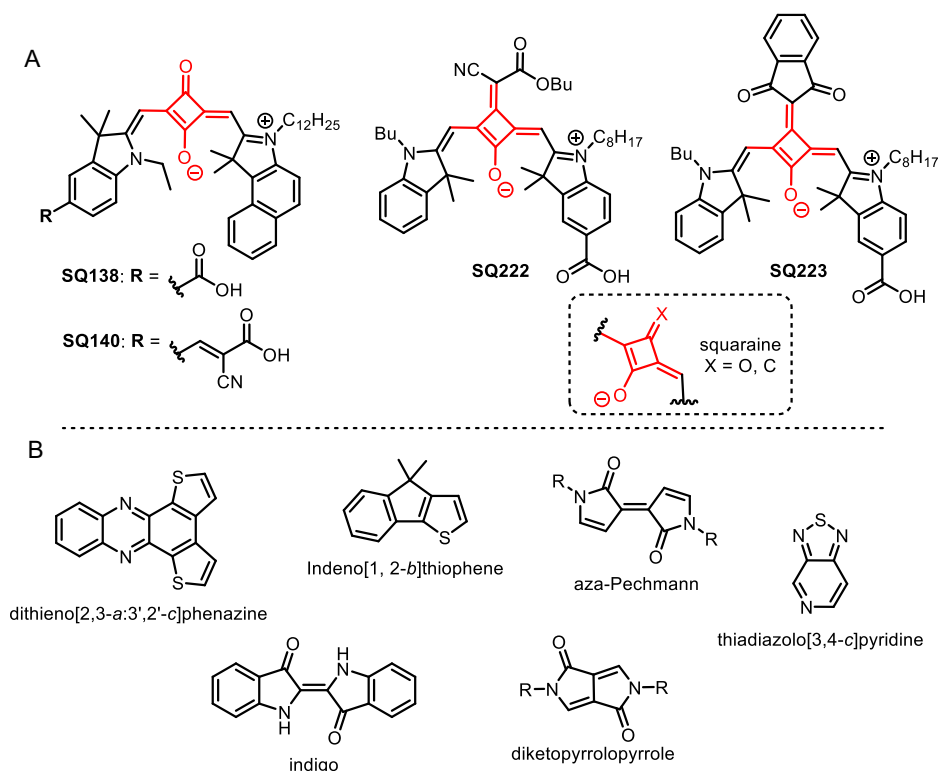


FIGURE 5 | (A) squaraine-based dyes **SQ138**, **SQ140**, **SQ222**, and **SQ223** and (B) several scaffolds exploited for green- and blue-colored organic fluorophores for NIR absorbing DSSCs.

also in D- π -A type DSSC [122–124] and p-type DSSCs [125]), the thienyl-BODIPY motif [126], or the thiadiazolo-pyridine structure (Figure 5B) [127]. It is worth pointing out that transparency is not always evaluated, and some dyes (especially the blue ones), despite the perceived color, display consistent absorption in the 500–600 nm region, a feature that does not ensure transparency in the final device.

3.3 | Near-Infrared Sensitizers

The NIR region (above 700 nm) represents an exploitable portion of the solar spectrum that could be exploited for light absorption for transparent DSSC fabrication purposes. The extension of the light absorption ranges from visible toward the NIR spectrum has been widely studied and considered as a guideline in dye design [128]. At the structural level, this has led to 1) the choice of an electronic push-pull (D- π -A) scaffold, 2) bandgap manipulation by judicious schemes of substitution with groups having a peculiar electronic effect, 3) diminution of conformational freedom with consequent augmentation of rigidity in the structure, and 4) steric protection.

In the field of BIPV DSSC, the exploitation of NIR with little to no absorption in the visible range is highly desirable to develop properly colorless devices that feature high AVT value together with quality color rendering. Given this appealing property, selective NIR sensitizers for DSSC were recently in the spotlight and were thoroughly reviewed by Grifoni et al. [42] Squaraine-based dyes [129, 130] were the first and most explored NIR selective sensitizers also for p-type DSSC [131–133], but despite tuning of π -bridges and anchoring groups [134], their absorption is still

tailing off down to 600 nm, making the dyes slightly green [135]. This issue was tackled by Galliano et al. by exploiting centrally symmetric benzindolenine squaraine dyes [136]. Upon modulation of the substitution pattern on the squaric core unit and the extension of the π -conjugation, they developed **VG12-C8** (Figure 6-left), whose absorption at 650 nm is relatively low but still reaches PCE of 4.6%. The same group recently developed a symmetrical heptamethine cyanine structure **VG20-C16** (Figure 6-right) with a strong and sharp S0–S1 transition in the NIR region ($\lambda = 832$ nm) and a minimized S0–S_n contribution in the far blue, where the human retina is poorly sensitive [137]. Due to a high aggregation issue, a dedicated sensitizing procedure was developed for **VG20-C16** to obtain a fully transparent and colorless DSSC that under AM 1.5G irradiation scored PCE of 3.1% with a scattering layer and 2.4% without it, AVT of 76%, and CRI of 92.1%, when associated with Co(bpy)₃ (3+/2+) as redox mediator.

More recently, Odobel et al. [138], reported pyrrolopyrrole cyanine derivatives as an alternative to the strongly aggregating NIR-absorbing cyanine, developing **TB207** sensitizer for colorless DSSC. The device scored a PCE of 2.5% with an AVT of 76% and a CRI of 93. Later, the same group reported **TB202**, demonstrating how the replacement of the phenyl ring on the bispyrrole core of **TB207** with a thiophene leads to a bathochromic shift of the absorption band (Figure 7) [139, 140]. Such a replacement of the benzene ring with heteroaromatic thiophene led **TB202** to display very low absorption below 700 nm and rendered it transparent in the visible region. In fact, in passing from **TB207** to **TB202**, the improvement of the transparency features (AVT passed from 63% to 67%) was not accompanied by an increase in PCE in the corresponding DSSCs (3.8 vs. 1.5%). This was

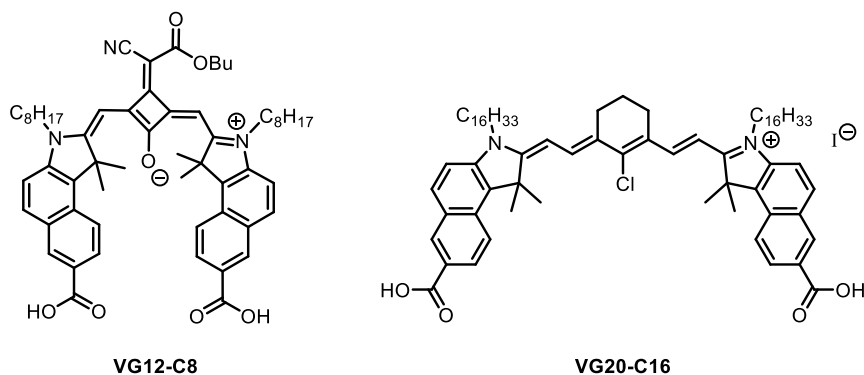


FIGURE 6 | Two NIR sensitizers for DSSC application. Left: squaraine-based fluorophore **VG12-C8**, and right: the symmetrical heptamethine cyanine dye **VG20-C16**.

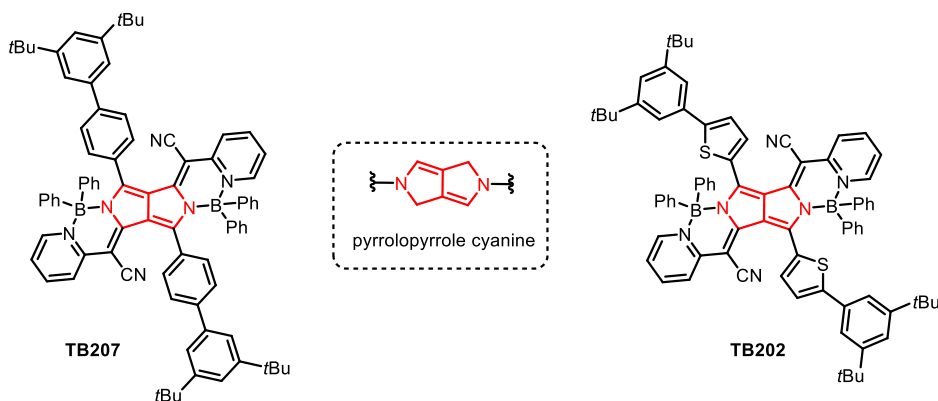


FIGURE 7 | Two pyrrolopyrrole cyanine-based NIR sensitizers for DSSCs: **TB207** (left) and **TB202** (right).

ascribed to a stronger aggregation tendency for the **TB202** dye in the immobilized state versus **TB207**, with a corresponding higher quenching rate of the excited state for **TB202** versus **TB207**, a slower kinetics of electron injection due to the lower LUMO level of **TB202** with respect to **TB207**, and a larger electron recombination rate in **TB202**-sensitized TiO_2 versus the **TB207**-sensitized version.

As a future perspective, new promising scaffolds have been reported in recent years based on tunable xanthene-based [141] and aza-BODIPY-containing [142] compounds that exhibit NIR absorption, implying their possible applicability for transparent PV applications.

3.4 | Advances on Dyes for Transparent DSSC

Cosensitization in DSSCs technology consists in anchoring a mixture of two or more different dyes onto the surface of the nanostructured photoelectrode [88, 143]. The main motivation of such a multidye approach is the broadening of the spectrum of absorption for the multi-dye-sensitized electrode with respect to the photoelectrode sensitized by a single dye in order to improve the overall conversion efficiency and achieve a panchromatic effect with consequent enhancement of the solar cell conversion efficiency. The multidye strategy in DSSCs can suffer of several limits: competitive absorption in case the spectra of the different anchored dyes present overlap in their absorption spectra (either

in the free, unbound state or in the anchored state); intermolecular charge transfer and consequent loss of the efficiency in terms of absorbed photon-to-current conversion (this term is dependent on the relative positions of HOMO and LUMO levels for the different dyes); intermolecular aggregation phenomena that alter the position of the relevant frontier electronic levels and the relative dynamics of electronic excitation/transfer with resulting quenching of excitation and loss of photon harvesting when strong coupling occurs between adjacent anchored dyes; partial loss of conformational freedom and lack of geometry optimization for the light-absorbing complex with resulting slowing down of the charge transfer process across the dye/electrode interface. Another factor of limitation is the competitive adsorption between different dyes on the basis of their diverse anchoring properties: this phenomenon occurs if one dye binds more strongly to the nanostructured electrode surface with respect to the other(s), with consequent starvation of anchoring sites for the weaker bounded dye(s). Moreover, the ratio control of different anchored dye is generally imprecise and difficult to achieve since the actual surface composition of the sensitized electrode does not coincide with the composition of the cocktail bath in which the electrode is immersed with resulting actual poor control over final surface ratios. Phenomena of desorption can be also favored in case of multidye sensitization since certain combinations of chemical components can trigger mutual dye displacement or dissolution off the nanostructured semiconductor matrix. As already outlined, cosensitization is the most commonly used strategy to improve the photovoltaic performances of a DSSC [88, 144], obtaining

panchromatic devices with enhanced photocurrent and wide IPCE, provided that every dye-sensitizer has a specific site of adsorption on the SC surface, and all sites are saturated after the double step of sensitization and cosensitization. In the field of transparent PV, this is achieved by pursuing high-transparency and low-efficiency IPCE in the window around 500–600 nm. Such an expedient has been applied to many dyes already reported in the previous sections [115, 123, 124, 126, 130, 145–147], but the most interesting reports in terms of transparency were obtained by cosensitizing UV- and NIR-selective sensitizers. One example is the **Y1/HSQ5** cocktail dyes [43] that, under AM 1.5G simulated solar irradiation, achieved a PCE of 3.66%, significantly higher than those of **Y1** (2.16%) and **HSQ5** (2.30%) single-dye cells [148] (Figure 8).

The improvement was mostly due to an increase in photocurrent while preserving a good AVT of 60.3% despite a low CRI, not assessed but deducible from the pictures of the devices. Such an achievement resulted from tailing absorption of **Y1** in the blue region and **HSQ5** in the red one. The adopted approach creates room for further improvements through the combination of dyes with selective absorption. Another fast-growing technology for transparent DSSC that relies on the development of advanced sensitizers is the energy management through dynamic color switching obtained with photo-switchable [149] (and eventually also electrochromic [150]) dyes. In this ambit, the DSSC's sensitizers capable of undergoing photoactivated reversible transformation largely affect the dye absorption profile too. The equilibrium between a high-transparency state and a more colored and photoactive one is generally driven by UV light photoactivation, while the reverse reaction is, in turn, thermally driven. The group of Demadrille et al. [151] exploited the D- π -conjugated-bridge-A structure, incorporating in the π -conjugated bridge a photochromic naphthopyran unit and modulating the donor part. The series of dyes **SF1-5** shows excellent photochromic properties and good photovoltaic properties too, establishing also the state of the art for this field (PCE up to 4.17%) (Figure 9A) [152].

Very recent work highlights cosensitization as an anti-aggregation and color-management tool in semitransparent devices. For example, cosensitizing NIR squaraine dyes with visible dyes that act as deaggregating agents improved IPCE and output: a D35:SQ258 mixture (1:5 molar ratio) reached 3.3% efficiency, rising to 3.5% with a cobalt electrolyte, while suppressing squaraine aggregation, evidenced by transient spectroscopy [153]. From a perspective standpoint, progress now hinges on quantitative control of mixed-monolayer composition (dye ratios, loading in nmol cm⁻², and displacement tests), using orthogonal

anchoring groups and preadsorbents, approaches already validated in record opaque cells to meet the multiobjective targets of PCE, AVT, CRI, and stability in BIPV.

Furthermore, in the **NP1-6** (Figure 9B) series, the modification in the naphthopyran substituents led to better discoloration kinetics, with cosensitization of photochromic dyes leading to PCE up to 4.34% [154]. The reduced number of molecular scaffolds reported so far for photo-switchable sensitizers in DSSC proves the difficulty to achieve both PV output and a large change in light transmission, a difficulty that is easily recognized as the principal next challenge in the design of dyes for transparent and smart DSSCs.

4 | Electrode and Interface Issues

Among other factors related to the chemical-physical stability of single components, the performance and long-term reliability of DSSCs are critically influenced by the quality of interfaces created by the electrodes with the electrolyte, with the redox mediator, and, in the case of the nanostructured SC electrode, by the interface existing between the SC and the chemisorbed sensitizer [155]. In addition to the interfaces just reported, it must also be considered the interface between the SC electrode and the transparent conducting substrate through which charge collection occurs [156–159]. Interface engineering directly affects charge injection and collection as well as unwanted charge recombination processes [160]. Therefore, interface engineering has a profound impact on the resulting photovoltaic performance of the device. In both n-type (e.g., with TiO₂-based anodes) and p-type (e.g., with NiO-based cathodes) DSSCs, a careful control over interfacial chemistry and morphology is essential for optimizing device efficiency and improving the operational temporal stability. In DSSCs, the imperfections such as atomic step edges, line defects, and oxygen vacancies, that are present on nanostructured TiO₂ or NiO electrodes act as undesired recombination centers. By “healing” these active sites, photoinjected electrons are driven toward the external circuit rather than being intercepted by the redox species in the electrolyte, thereby improving the overall PCE of the device [161, 162]. In the process of nanoelectrodes preparation for DSSCs the researchers adopt specific interface engineering techniques like TiCl₄ treatment of the nanostructured photoanode, deposition of ultrathin inorganic coatings (such as MgO, ZrO₂, or Al₂O₃) with passivating properties onto the photoelectrodes, and chemical paths with acid agents to minimize undesired surface and edge defects that provoke localized electronic and chemical phenomena at the boundaries of the nanoelectrode [163]. These surface/interface engineering strategies passivate oxygen vacancies, render more uniform the electrode surface, and suppress charge recombination (back electron transfer) with favorable effects on the resulting efficiency of light conversion. Surface and edge defects are mainly due to the nonuniformity of dye loading and to the presence of dangling bonds at the outer surface of the electrode for the unsaturation of the valence of surface atoms. Nonuniform dye-loading can create zones of lower efficiency of photo-induced charge injection, whereas unsaturation favors charge recombination and introduces catalytic activity with detrimental effect on redox-active species impeding the correct operation of charge transfer [164–166].

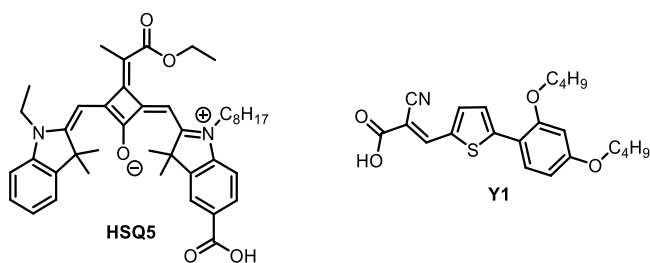


FIGURE 8 | Chemical structures of the two compounds employed in the cocktail dyes for cosensitization of DSSC photoanodes.

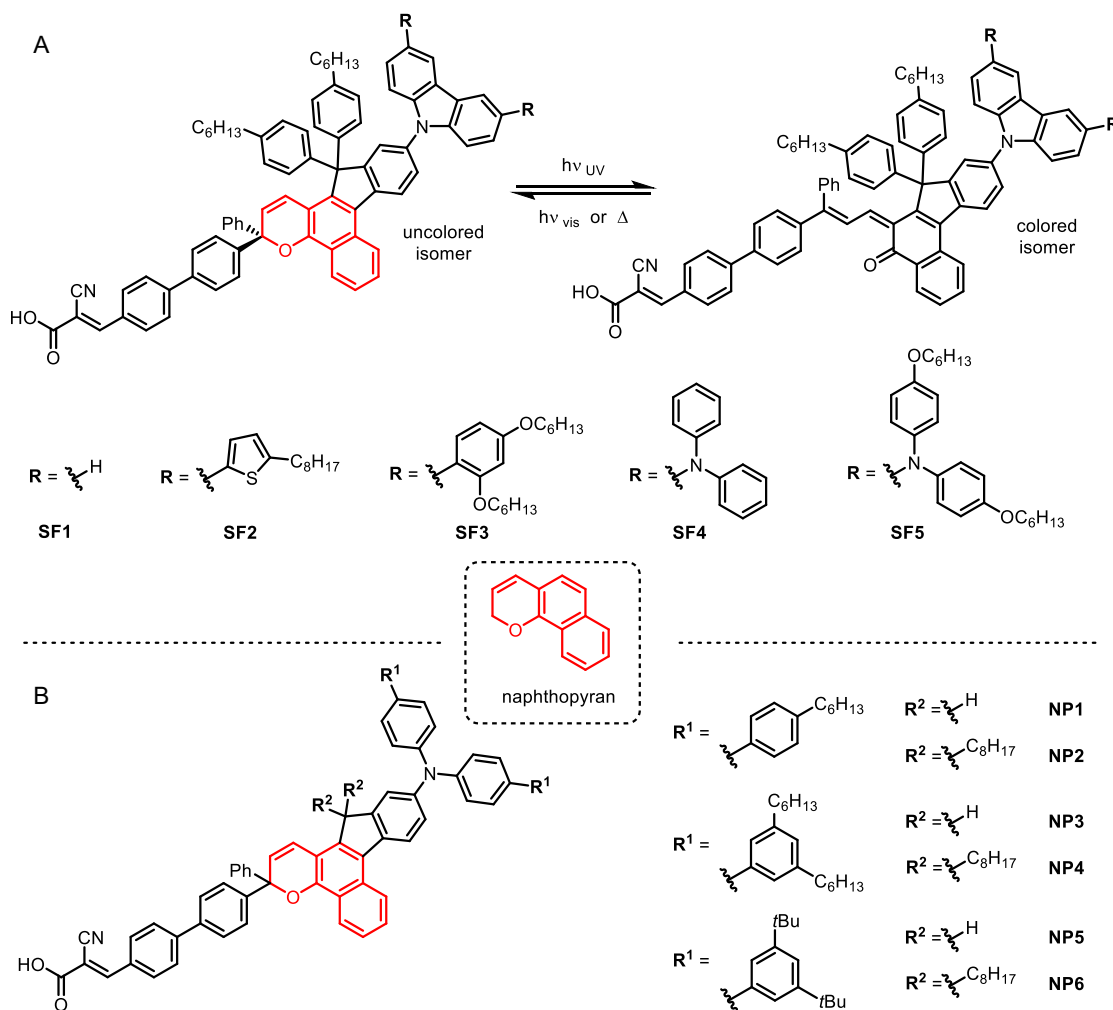


FIGURE 9 | Naphthopyran-based fluorophores and their photochromic behavior: (A) the SF1-5 series, and (B) the NP1-6 series.

In a DSSC, the mechanism of charge transport is governed by:

1. Trap-limited diffusion of electronic charge carriers (electrons in photoanodes and holes in photocathodes) through the nanostructured photoelectrode.
2. Back-donation phenomena at photoelectrode/electrolyte interface caused by a relatively slow diffusivity of the photoinjected electronic charge carriers (especially at photocathodes) that live sufficiently long to interact with the photo-redox product in the electrolyte.
3. Diffusivity of the redox species and ionic charge carriers in the electrolyte.

Therefore, the charge-transport mechanism in DSSCs appears as a complex interplay between electronic charge diffusion through a nanostructured/mesoporous semiconducting electrode, ionic diffusion through the electrolyte, and an interfacial charge transfer driven by photo-driven (fast) changes in electronic states of immobilized molecular sensitizers and of the nanoelectrode surface. In this context, the nature of transport is mainly of diffusive type, whereas built-on electric fields play a less important role in DSSCs with respect to junction-based photovoltaics systems. The parameters governing the preparation and the processing of nanostructured electrodes for DSSCs (thickness, nanoporosity,

substrate adhesion, dye-loading capacity, geometric area, and degree of opacity) [167] affect mostly the dynamics of charge motion through the bulk and the electrode/electrolyte interface rather than influencing the corresponding microscopic mechanisms of transfer. On the other hand, the parameters of nanoelectrode preparation have a direct influence on the effectiveness of interfacial charge collection/injection and must account for the minimization of charge recombination at the nanoelectrode/electrolyte interface and charge trapping within the nanophase. Processing parameters of the nanoelectrodes define the path that the electronic carriers have to navigate through the electrode and the rate at which the electronic carriers are exchanged at the different interfaces the nanoelectrode forms in a DSSC. Modifying these parameters fundamentally leads to the variations of the efficiency with which the electronic charge diffuses in the nanostructured electrode prior to recombination/trapping. In the specific, the sintering temperature, the ramp of its variation, and the duration of heating all affect the process of nanoparticles necking, substrate adhesion, degree of porosity, and the formation of the charge percolation path. A too-short process of sintering leaves morphological boundaries that can favor increase electron trapping phenomena. Conversely, a too-long process of sintering can lead to particle aggregation (over-sintering effect), which minimizes internal surface area, stifles subsequent dye loading, and reduces the interfacial active

area for the purpose of charge transfer. Temperature processing parameters determine the porosity of the nanostructured electrode, and a balance must be achieved considering that high porosity warrants the adequate infiltration of the liquid electrolyte containing the redox couple. In case the pores are too small (possible consequence of over-sintering), problems of wettability might verify with diminution of the effective contact area between electrode and the electrolyte and consequent reduction of photocurrent density. When porosity is excessively high, problems of necking might occur at the interface formed by the nanostructured electrode and collector surface with the consequent diminution of charge collection efficiency, which brings about the reduction of the photocurrent density. Electrode thickness, a parameter depending on mass electrode and the density of the constituting materials in the nanostructured version, is chosen in a way that warrants low intrinsic optical absorption to attenuate self-absorption, that is, a phenomenon is unhelpful for the process of charge injection in DSSCs and assures a path for the finalization of charge collection, which needs to be longer than the diffusion length of the electronic charge carriers within that electrode. When too thick electrodes of very transparent materials are employed, the resulting light-harvesting properties would be certainly favored, but problems of recombination might occur for the too-long path of collection with respect to the diffusion length. Use of very transparent materials as photoelectrodes for DSSCs should warrant a value of lifetime for the respective electronic charge carriers, which is longer than the recombination time.

The contact electrical resistance at the photoelectrode/charge collector interface and at the junction formed by the charge collector with external circuit contact are scarcely influential on charge transfer mechanism in DSSCs since all the factors governing the DSSC kinetics exert their influence in steps that phenomenologically precede both charge injection through the photoelectrode/charge collector interface and the charge transfer through the junction collector/contact resistor. Electronic effects in these terms of contact resistance for DSSCs can be of blocking type if the process of charge extraction from one phase to another is governed by the energy scheme of Schottky-type junction, which refers to conductive materials of different nature (metal-semiconductor the latter being subject to a variable extent of doping). On the other hand, high contact resistances generate large Ohmic losses that negatively affect negatively the current density through the external circuit and lower the resulting output of electrical power with consequent diminution of the conversion efficiency.

Optical scattering is a phenomenon that can be introduced in DSSCs for optimizing the process of light utilization [168]. With the insertion of scattering agents in the photoactive layer, which can be either electrically active or not, the efficacy with which the light is absorbed rises thanks to the reflecting/diffusing nature of these agents toward photons. This is originated by the increase of the traveling distance for the photons inside the photoelectrode with the photons that are back reflected toward the sites where dye-sensitizer molecules are chemisorbed. As a consequence of these photon transport phenomena, there is the enhancement of the amount of absorbed light, which favors photo-charge generation with consequent increase of PCE [169]. The employment of scattering agents in DSSCs improves the

absorbed photon-to-current efficiency through the step of photo-injection with an increase of photocurrent due to the augmentation of the photogenerated electronic charge carriers, but the use of optical scatterers in DSSCs does not favor directly the mechanisms of charge transfer.

The scaling-up of the single DSSC from a small to a larger area increase renders increasingly important the lateral electrical resistance since electronic charge carriers must travel longer lateral distances along the charge collector to reach the metal finger contacts or interconnects. Since charge-collecting materials (namely, TCO sheets) have a nonnegligible sheet resistance, the involvement of longer paths for the electronic charge carriers drastically brings about an increase of the total internal resistance with negative influence on DSSC PCE (diminution of current density for Ohmic losses). Moreover, the expansion of the DSSC area can lead to in a nonuniform current distribution with generation of surface distribution of potential drop due also to the more probable presence of point defects that can increase the probability of charge trapping. It is then expected that an increase of DSSC area will lead to the increase of the current value but not on the current density, that is, the term referred per unit area. In consideration of the influence the DSSC area can exert on charge transport mechanism, it is reasonable to consider primarily the increase of the probability with which charge trapping phenomena can occur in larger area DSSCs for the higher probability of introducing impurities in the phase of paste spreading (nanostructured electrodes precursors) and in the step of charge collector deposition onto the supporting substrates.

4.1 | Stability of TiO₂ and NiO Interfaces

Nanostructured TiO₂ with an anatase crystal structure is the most widely employed photoanode material due to its suitable conduction band position, high surface area, and chemical stability [170, 171]. However, under operational conditions, especially prolonged UV exposure, TiO₂ can catalyze photodegradation reactions, particularly in the presence of oxygen and moisture. Surface trap states—originating from oxygen vacancies, under-coordinated Ti atoms, and surface hydroxyl groups—can act as recombination centers and impede electron transport [172, 173]. In addition, weak dye anchoring or dye desorption from the TiO₂ surface can further deteriorate the PV performance [174, 175]. A sketch of the surface trapping and of the recombination pathways is given in Figure 10.

In p-type DSSCs, NiO is the benchmark material for the preparation of nanostructured photocathode: it generally suffers from high self-optical absorption (a characteristic that obliges operators to adopt thinner nanostructured films with respect to the less optically thick TiO₂), it has tendency to give charge recombination with I₃[−]/I[−] redox couple, and it displays low hole mobility. The oxidation of Ni²⁺ to Ni³⁺ represents a p-doping process with concomitant uptake of an anion and formation of nickel hydroxides on NiO surface if the oxidation is carried out electrochemically and occurs in aqueous or humid conditions [69]. Differently, if the electrochemical oxidation of NiO is carried out in nonaqueous solvents, the uptake of the electrolyte anion will occur [68, 70, 72]. The oxidation of Ni²⁺ → Ni³⁺ is consequent to a chemical process

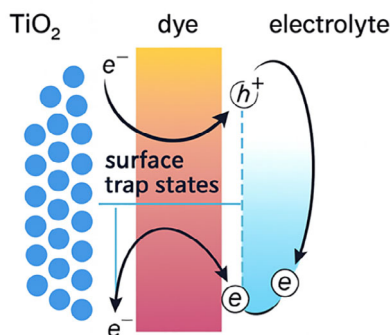


FIGURE 10 | Schematic illustration of surface trap states and recombination pathways at the TiO_2 /dye/electrolyte interface that is present in DSSCs.

involving an oxidizing agent like O_2 ; because of that, the uptake of O^{2-} anion will simultaneously occur. Once oxidized, the resulting nonstoichiometric nickel oxide will present irreversible changes in surface chemical composition, displaying an increase in charge carrier density, shifts in frontier energy levels, and introduction of trap states. These alterations hinder hole injection from the dye and increase interfacial resistance. Strategies such as surface doping (e.g., with isovalent Mg^{2+} or aliovalent Li^+) [176–179] and postsynthetic annealing have been explored to improve NiO crystallinity and reduce surface defects [180, 181].

4.2 | Charge Recombination and Passivation Strategies

A major loss pathway in DSSCs involves recombination of photo-injected electrons in TiO_2 with oxidized species in the electrolyte (or holes photo-injected in NiO, which recombine with reduced species) [182–186]. For TiO_2 -based systems, recombination with triiodide (I_3^-) or cobalt(III) complexes limits the open-circuit voltage (V_{oc}) and fill factor (FF). This issue becomes more critical in thin-film DSSCs designed for transparency or indoor applications, where the limited thickness combined with large surface areas of the photoelectrodes increases electron diffusion constraints. Passivation strategies include inorganic oxide coatings, where thin layers of wide bandgap materials such as Al_2O_3 , ZrO_2 , or MgO can be deposited onto TiO_2 to reduce surface recombination without severely blocking electron injection [164, 187]. Use of ZrO_2 as charge recombination quencher has also been proposed for p-DSSCs [54]. An alternative to the addition of insulating oxide layers (or in the form of dispersed nanoparticles) is the adsorption of self-assembled monolayers: molecules like chenodeoxycholic acid (CDCA) are coadsorbed with the dye to reduce the surface density of trap states, prevent dye aggregation, and promote better dye orientation for interfacial charge transfer and intramolecular charge transport [133, 188–190]. At the technological level, an advanced method of electrode surface passivation is represented by atomic layer deposition technique that allows conformal coating of the semiconductor surface with sub-nanometer precision, enabling stable interfacial passivation layers that reduce recombination and enhance charge separation [191–194]. A sketch of the passivation-dependent recombination pathway is given in Figure 11.

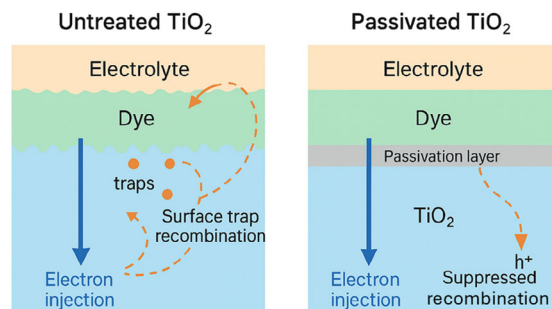


FIGURE 11 | Recombination pathways in untreated versus passivated TiO_2 electrodes for DSSC applications.

4.3 | Surface Engineering and Blocking Layers

Interface engineering plays a vital role in determining the efficiency and durability of DSSCs. Among the various strategies developed to optimize interfacial charge transfer and minimize energy losses, the application of blocking layers and surface functionalization stands out as particularly effective [195–197]. A commonly employed method to reduce charge recombination at the photoanode is the deposition of a compact TiO_2 layer between the transparent conductive oxide substrate, typically fluorine-doped tin oxide (FTO), and the mesoporous TiO_2 layer [198–201]. This dense film acts as a physical and energetic barrier that prevents the undesirable back-electron transfer from FTO to the oxidized species in the electrolyte. As a result, V_{oc} increases, and the overall photoconversion efficiency improves. Additionally, by limiting direct contact between FTO and the electrolyte, the blocking layer contributes to improved operational stability, which is particularly beneficial under prolonged light soaking or in thermally stressful conditions.

In parallel, surface functionalization of the mesoporous TiO_2 photoanode provides additional opportunities for performance enhancement. Introducing chemical groups such as silanes, phosphonic acids, or molecular linkers on the TiO_2 surface strengthens the binding of dye molecules, reduces surface trap densities, and modulates the alignment of energy levels between the dye and the semiconductor [202–207]. This results in more efficient electron injection and better charge separation. These functional groups can also prevent dye aggregation, increase chemical robustness, and influence interfacial wettability—all factors that collectively affect both the initial performance and long-term stability of the device.

In the case of p-type DSSCs, where NiO is used as the photocathode, similar principles apply, albeit with distinct challenges. NiO typically suffers from low hole mobility and a high density of surface states that promote recombination [208–212]. Surface engineering approaches such as passivation layers, anchoring groups with electron-withdrawing functionalities, and the incorporation of protective overlayers have shown promise in enhancing hole injection efficiency from the excited dye and suppressing recombination at the NiO/electrolyte interface [55, 213–215].

Meanwhile, the development of robust and cost-effective CE materials has become a major focus in DSSC research. While platinum remains the standard due to its excellent catalytic

activity, especially for the triiodide/iodide redox couple, it is expensive, scarce, and susceptible to detachment in the presence of alternative redox mediators like cobalt or copper complexes. This has led to the exploration of various alternative materials. Carbon-based materials, such as carbon nanotubes, graphene, and activated carbon [216–219], offer high surface areas and good electrical conductivity, making them suitable for catalyzing redox reactions at the CE. In addition, transition-metal dichalcogenides (TMDs), including molybdenum disulfide (MoS_2) [220, 221] and tungsten disulfide (WS_2) [222–224], have attracted attention due to their layered structures, large surface areas, and promising electrocatalytic behavior.

Conducting polymers such as PEDOT:PSS [225] and polyaniline [226, 227] also present viable CE alternatives for DSSCs since these systems offer flexibility, tunable electronic properties, and relatively straightforward fabrication processes. Moreover, these materials exhibit good electrocatalytic activity toward a variety of redox mediators [228–230] and can be processed on flexible substrates, making them attractive for lightweight or wearable DSSC applications [231].

Taken together, these surface and interface modifications—ranging from photoanode optimization to CE design—underscore the complexity and interdisciplinary nature of DSSC engineering. The interplay between surface chemistry, materials science, and device physics offers multiple levers through which efficiency and stability can be systematically improved. In Figure 12, two examples of TiO_2 blocking layer and carbon-based CE are shown [217, 232].

4.4 | Interfacial Dynamics and Lifetime Studies: How Characterization Techniques Evolve

Advanced characterization techniques such as electrochemical impedance spectroscopy [184, 233–235], transient photovoltage/photocurrent measurements also with pulsed light techniques [236–239], and intensity-modulated photocurrent/photovoltage spectroscopy (IMPS/IMVS) [240, 241] provide insights into charge transport and recombination kinetics in DSSCs. Time-resolved studies under accelerated aging conditions (e.g., thermal cycling, UV soaking, and humidity tests) are essential to identify degradation mechanisms and benchmark the interfacial stability in order to improve the durability of the DSSC [242–245]. In the ambit of the study of the kinetics governing the various transport phenomena in a DSSC, the advancements in established

characterization techniques, their combination for a multitechnique correlative analysis, the development of new characterization techniques, as well as the evolution of aging protocols, are fundamental to impart more meaningfulness to the outcomes and to increase the long-term predictability of the DSSC performance. In this evaluative process of the DSSC performance, the operator should consider the resource of the artificial intelligence (AI) for the availability of an increasing amount of data, which facilitates the comparability of many different DSSCs and renders easier the choice of the most appropriate photoconversion system in a given operative environment [246]. Besides the involvement of AI, the operator of DSSCs should also take into account the increasing level of automation reached in many experimental techniques of photoelectrochemical cell characterization. The latter aspect favors the development of self-driving laboratories [247] that can be installed also in a nonconventional ambient, for example, man-free, thus offering the possibility of realizing experiments of DSSC testing also under water or in space and, possibly, extending the use of DSSCs.

4.5 | General Considerations on the Reliability of DSSCs Data

In general, the reliability and the consistency of DSSCs data (reproducibility of $J-V$ profiles and, consequently, of the power output) are strictly related to the chemical-physical stability of the materials (nanostructured semiconductors, dye-sensitizer, electrical conductive coatings) and the corresponding interfaces, of their configuration (with particular attention to the deformability of the substrates under varied conditions of pressure and, more importantly, of temperature and the associated risks of delamination), and the chemical-physical stability of the auxiliary parts (electrical contacts and sealing materials) under the adopted conditions of illumination (nature/spectrum of the light source and intensity), electrical polarization, humidity, temperature and resulting power output during cell testing and characterization. Data reliability of DSSCs is primarily conditioned by the stability of the environmental conditions during the phases of cell testing and characterization, and more reliable have to be considered the data obtained from cell tests conducted in indoor conditions with respect to those carried out in outdoor conditions for obvious reasons of controllability [248, 249]. Imposed conditions of photoelectrochemical testing can intrinsically generate unreliable data for DSSCs: in case of the registration of $J-V$ curves and electrochemical impedance measurements, hysteresis phenomena can occur in dependence, respectively, of the sweep rate and sweep direction of the potential [250] and the frequency of potential oscillations that generate variable phases of impedance response [226]. Same considerations also apply for the occurrence of hysteresis phenomena detectable in IMPS and IMVS spectra of DSSCs [251]. The origin of photoelectrochemical response hysteresis in DSSCs originates from the slow mass transport of redox mediator in the DSSC electrolyte being the transport of the redox mediator under diffusion control and constituting the rate-determining step of the overall kinetics of DSSC dynamics [14, 252, 253]. In fact, charge photo-injection from the excited dye to the nanostructured semiconductor, the electronic charge transport through the semiconducting and metallic electrodes, and interfacial electronic charge transfer between the redox shuttle and the electrode in DSSCs all represent kinetic

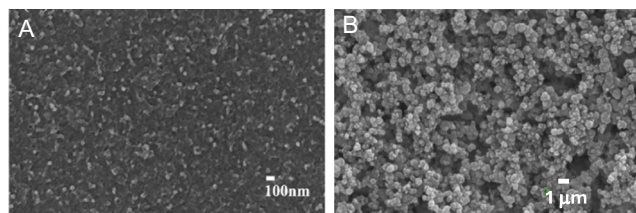


FIGURE 12 | SEM images of (A) TiO_2 blocking layers [reproduced with permission from ref. [232]]. Copyright 2021, MDPI. and (B) carbon-based counter electrodes, highlighting surface morphology and porosity [reproduced with permission from ref. [217]]. Copyright 2021, ACS.

steps that are faster than the transport of the redox shuttle through the electrolyte. The sluggishness of transport of the redox shuttle affects the initiation of the other elementary steps in DSSCs and can produce local accumulation of charges with generation of capacitive effects associated with localized electric fields [254].

At the basis of data reliability of DSSCs, there are also memory effects that can be observed during DSSC operation and characterization. These effects consist in photo-responses with transient character, which depend on the previous history of cell conditioning [either electrical (DSSC polarization in the dark), optical (DSSC illumination at controlled kinetics as in IMPS and IMVS experiments [251] and during LBIC recording [255] at different scan rates), or chemical (treatment of photoelectrode surfaces under specific conditions of sensitization) [256]] prior to illumination in steady conditions. In case of LBIC mapping of the photoelectrochemical activity of a DSSC, the point-by-point output of the electrical power in a DSSC results dependent on the conversion efficiency previously determined at adjacent photo-excited positions with consequent production of nonuniform images of photoelectrochemical activity, which lack of sharpness being subject to lateral photo-resistive effects [255]. These effects of memory are associable with the generally slower response to irradiance of a DSSC with respect to a solid-junction PV system due to the mass transport phenomena in the DSSC electrolyte, which control the overall kinetics of charge transfer. The memory effect related to chemical conditioning can occur in case of DSSCs employing large molecular sensitizers (e.g., zinc-porphyrin derivatives) [256]: at the preparative level, phenomena of sensitizers self-assembling can be verified during sensitization with the anchored sensitizers forming an oriented monolayer of sensitizers, the geometry and orientation of which are not dependent on the nature of the dipping solvent for a given specific zinc-porphyrin sensitizer. In this latter case, the memory effect is produced during the process of coloration/sensitization of the nanostructured semiconductor and consist in the retention of the intermolecular superficial arrangement due to the strong intermolecular interactions between adjacent chemisorbed sensitizers, with the surface orientation of the chemisorbed sensitizers not dependent on the nature of the dipping solvent.

The reproducibility of the electrical power output of DSSCs varies within a deviation range of 5%–10% when cell testing is conducted under controlled laboratory conditions or in an indoor ambient [257]. Deviations in power output can become considerably larger (up to 30%) when the DSSCs performances are recorded in outdoor operative conditions with aging effects that can severely affect DSSC durability also in the first phases of testing depending on the type of degradation, which is initiated at the start of DSSC operation [258]. At the basis of reproducibility in the conversion performance of DSSCs stands out crucially the difficulty in reproducing semiconducting nanomaterials with well-defined thickness and optical transmission, large surface area, interconnected pores for favoring rapid electrolyte permeation and increasing the interfacial contact area, defined crystalline phase(s), intrinsic and interfacial chemical-physical stability, and optimized chemical surface composition for a reproducible process of sensitization. In addition, for the improvement of DSSC performance reproducibility, the utilization of sensitizers and redox mediators with optical, photo-physical/chemical,

electrical, and electrochemical stability is of capital importance. A reduced volatility of the electrolytic solvent is another important factor that can favor DSSC performance reproducibility with prevention of leaking/evaporation effects. At the module level, the scaling of the photoactive and the electric conducting areas in DSSCs can bring about uncontrolled gradients of resistance in larger area devices with risks of increase for the variation coefficient of the photogenerated power output [259].

The efforts in achieving reliability for a given set of data/experiments from/on DSSCs are necessarily preceded by the preliminary recording of an extremely large amount of DSSCs data referring to the significant indicators of power conversion performance. Such DSSC indicators could have been acquired under quite different experimental conditions (operative, simulated, accelerated, with control of one or more stressors or other), and the consequent availability of large sets of data has offered the opportunity of conducting a meaningful study in DSSCs at the statistical level involving also advanced tools of data processing, for example, the artificial intelligence (AI) (vide infra). In fact, the statistical analysis of DSSC data comprehends the collection of large amounts of experimental values of the relevant parameters of cell conversion efficiency: short-circuit current density, open-circuit voltage, maximum power point, fill factor, absorbed photon-to-current conversion efficiency, incident photon-to-current conversion efficiency, and overall PCE, which are all dependent on each other by definition. Together with these performance metrics, the statistical analysis of DSSCs also considers the parameters of durability [260], which are determined through the monitoring of the performance metrics under predefined conditions of cell aging. The durability parameters consist in the determination of DSSCs lifespan and degradation rate [104], which are both primarily addressable to the thermal stability, UV resistance, and electrolyte volume/mass retention in DSSCs. With this setting, the durability parameters allow the direct evaluation of the sealing integrity, solvent volatility, dye desorption rates, and electrode corrosion resistance. Performance and durability metrics are both groups of key analytical metrics in the ambit of DSSCs, and the analysis of their temporal trends allows the evaluation of the actual progress in the processes of DSSCs fabrication. The monitoring and quantification activities related with the statistical analysis of the key metrics of DSSCs pursue the finality of orientate the future decisions in the design of the process of DSSC fabrication and the choice of the materials. Methods in use for the statistical analysis of DSSCs are:

1. **Descriptive Statistics:** It uses the approach of the batch experiments with the calculation of the maximum, minimum, mean values, and the corresponding standard deviation of the DSSC PCE from groups of experiments differing for specific conditions of materials preparation, module area, cell number in module, conditions of cell assembly, dye type, nature of the nanostructured electrode (morphology, crystalline phase), number of layers in the electrode, type of redox shuttle, type of additives in the electrode and in the electrolyte, and processes of electrode cleaning. A particularly useful aspect of this statistical approach is related to the possibility of correlating diverse metrics of interest, as in case of the work presented in ref. [261] in which charge transport/transfer kinetics and dye electronic properties of DSSCs are confronted in a unified view. An

example of descriptive statistical analysis of DSSCs is reported in ref. [262].

2. **Analysis of Variance:** It is a statistical approach that compares studies and analyses results within the same group of experiments (it would correspond to a single group of the descriptive statistics approach), focusing on a specific aspect and its influence on DSSCs performance. It is particularly useful in evaluating if controlled variations of fabrication conditions result in statistically meaningful differences in DSSC conversion efficiency. An example of analysis of variance for DSSCs in terms of the nature of the dye-sensitizer is reported in ref. [263] with presentation of a meta-analysis approach.
3. **Analysis of Time-series:** In the ambit of the statistical analysis, the time-series analysis method studies data points collected over specific time intervals in order to recognize temporal patterns like trends, cycles, and seasonal changes with the purpose of forecasting future events (predictive function). In this analysis, the individuation of noise due to random and unpredictable variations in the data is also taken into account. In the particular case of DSSC conversion performance, the time-series approach is applied to track the degradation of the performance over time for a given DSSC device monitoring temporally specific indicators of degradation that are not derived from the direct measurement of electrical parameters, namely, dye (or redox shuttle) photobleaching, electrolyte evaporation, loss of mass, and changes in volume/shape. Data were timestamped with a digital recording system in order to be successively processed and analyzed with opportune algorithms. Therefore, the approach of time-series analysis has not solely a monitoring purpose but is also employed for predictive analysis and modeling in the successive phases of data analysis/processing, which are successive to initial phase of data collection. In this perspective, the time-series analysis approach constitutes a method which is prone to automatization for the generation of an output with the time as an independent variable. The time-series analysis can be a multivariate method with the collection of data from multiple variables. In the work reported in ref. [264] in which the authors have employed statistical time-series modeling with multivariate analysis for forecasting the long-term degradation of the performance of power conversion in a DSSC.

The availability of large amounts of data on DSSC conversion performance allows the use of AI for automating data analysis taking eventually into account the data previously correlated through experiments or computational methods. The exploitation of machine learning tools [265–267] allows AI to predict DSSCs conversion efficiency [268] and degradation times [269] and to design materials [270, 271] for the fabrication of DSSCs with expectedly improved behavior [272]. Such a preparatory set based on AI is beneficial in reducing drastically the efforts spent in trial-and-error experiments on DSSCs with consequent diminution of costs and times of test realization.

AI output is then based on large, multiparameter datasets, and its models allow both temporal and spatial mapping of critical variables, for example, Temperature of operation and processing, dye type, nanostructured electrode chemical-physical characteristics, and contact positioning among

others—to temporal PCE losses. These possibilities offered by AI in the field of DSSC analysis allow researchers to estimate accurately the operational longevity of the DSSCs without waiting long times for real or accelerated tests of cell aging. The algorithms trained by AI for use in DSSCs analysis are briefly mentioned hereafter since a detailed description of these algorithms goes beyond the scope of the present review. The most appropriate algorithms for DSSCs analysis are long short-term memory model [269] for mapping time-dependent curves of decay and the models artificial neural network [268], random forest [273], and K-nearest neighbor [274] for the individuation of the physical parameters that constitute the dominant drivers of early-stage degradation (for example, in case of the nanostructured electrode, the individuated drivers are the natures of precursors and substrate, the temperature of deposition, the chemical composition of the ambient of deposition, and thin-film thickness).

5 | Transparent DSSCs: Issues

As anticipated in the precedent sections, the most important pursue inspiring the present research on DSSCs for extending their use in an evolving context of energy consumption [246] is the achievement of transparent devices that are capable to produce electrical energy in a passive fashion, that is, requiring only a renewable source of energy input while maintaining visual comfort in the ambient DSSCs delimitate [145, 275–278]. In this context, also the aesthetic feature of these light-conversion devices [42] constitutes an issue in itself since transparent DSSCs are expected to carry out their function where humans reside. The relevant ambits of transparent DSSCs are BIPV, AIPV, and indoor applications [49, 279–282]. For agrivoltaics application, vide infra. The parameter of AVT becomes central in the research on DSSCs for BIPV (or AIPV) and for a hypothetical DSSC with 100% of AVT is predicted a PCE of 21% [283]. The study also reports the comparison with the theoretical upper limit of PCE when the PV device is opaque and for which is 33%, that is, the Shockley–Queisser limit [284]. Transparent and colorless DSSCs with an AVT higher than 75% (Figure 13) present a maximum



FIGURE 13 | Picture of the semitransparent DSSC employing I_3^-/I^- as redox couple and a near infrared sensitizer based on polymethine cyanine structure. The resulting AVT is 76% with overall PCE of 3.1%. Reproduced with permission from ref. [137]. Copyright 2021, Wiley.

PCE of 3.1% [137], a value that is of an order of magnitude lower than the theoretical limit.

This important result of Naim et al. [137], stimulates further work in the field of transparent DSSCs, with researchers now aiming at reaching a superior performance of conversion, which will approach 10% for a device with an AVT of 75%. Depending on the device destination, a trade-off between efficiency and transparency is required. A handy rule of thumb is that the product PCE x AVT should be not less than 450 for a transparent DSSC worth of practical consideration. The product PCE x AVT has been denominated LUE, and it has been recently defined for the emergence of highly transparent photovoltaics for distributed utilization, that is, for a type of utilization that is characterized by the vicinity of solar-powered devices to the site of electricity consumption [285]. The concept of distributed utilization of solar cells—useful in BIPV, AIPV, and solar powering of portable and wearable electronics, involves small-scale PV devices and is somewhat in contraposition to the system based on the centralized distribution of the electrical power, a system the cons of which are the high costs of distribution and high losses in dispatching energy from the distribution center to the site of consumption. The application of the rule here proposed would predict that for a device with AVT = 75%, the PCE should be not less than 6%, while a PCE of 20% should be accompanied by an AVT value of at least 23%. In the competition toward the fulfillment of the rule of 450, transparent DSSCs will have to compete with perovskite solar cells [286–293], that is, the systems that represent the other protagonist in the context of transparent and quasi transparent HOIPV and organic PV cells [294]. In the pursuit of ensuring visual comfort in the spaces where DSSCs operate and humans reside, beside AVT, another parameter of interest is the index of color rendering (CRI) [295]. CRI is a parameter that refers to the quality of the vision, which is obtained with a given light source and has the maximum value of 100 when referring to daylight. CRI evaluation is useful in applications in which see-through is a feature that characterizes the device under consideration, and a high value of CRI in the ambient of vision confined by the see-through device is a desirable outcome. For DSSCs with visible light transparency, the determination of the corresponding CRI in the ambient of vision delimited by the DSSCs is an additional crucial aspect for evaluating the quality of the DSSC. The AVT of a PV device is directly determined by the transmission spectrum of the DSSC within the range of 380–780 nm, that is, the range that is bound to the spectral sensitivity of the human eye. For imparting an aesthetic significance to the photoconversion performance of transparent DSSCs, another parameter of transmittance including CRI (a light source-related parameter) and the photopic response (a human eye-related parameter) should be considered. At this regard, the photopic AVT, AVT_{ph} , (a dimensionless parameter), defined as

$$AVT_{ph} = \frac{\int_{380}^{780} \Phi_{src}(\lambda) * T_{PV}(\lambda) * PR(\lambda) d\lambda}{\int_{380}^{780} \Phi_{src}(\lambda) * PR(\lambda) d\lambda} \quad (1)$$

in which $\Phi_{src}(\lambda)$ is the wavelength dependent photon flux of a given light source (in $s^{-1} cm^{-2}$ —this is the term related to CRI and usually refers to the irradiance of the AM 1.5 G sun simulator), $T_{PV}(\lambda)$ is the wavelength dependent transmittance of the photovoltaic device, $PR(\lambda)$ is the wavelength dependent photopic

response of the human eye (term of perception), is a parameter considered as a figure of merit of the transparent PV device [296]. Another feature that is sought after in the recent DSSCs research is the realization of bifacial devices in which both sides of the cell can be photoactivated exploiting both direct light and reflected/diffused radiation [7] in order to increase the degree of utilization of the light for photoconversion purposes. For bifacial PV cells the parameter of interest is the ratio

$$[PCE_{rear}/PCE_{front}] * 100 \quad (2)$$

where $PCE_{rear/front}$ denotes the PCE at rear/front illumination. Bifacial photovoltaic technologies [297] are not necessarily a synonym of transparent photovoltaic system. In case of DSSCs with tandem configuration, that is, with two photoactive electrodes in the same electrochemical cell, the aim is to harvest photons at as many as possible different energies with the two photoelectrodes displaying complementary light absorption features and with illumination that can be bifacial (Figure 14) [53, 55–57, 59, 70].

In fact, the electrical output of such bifacial DSSCs is limited by the current density value achievable by the less performing photoelectrode, and irradiation geometry as well as the nature of the light sources have to be considered in the design and development of the bifacial tandem DSSCs [298, 299].

The achievement of DSSCs with transparent or semitransparent features has important repercussions also in agrivoltaics [300]. This is particularly evident when the replacement of the transparent walls/roofs of greenhouses with semitransparent solar cells is considered [301]. The approach introduced by the extended use of PV in agriculture is innovative because it considers a dual use of the land: for the simultaneous production

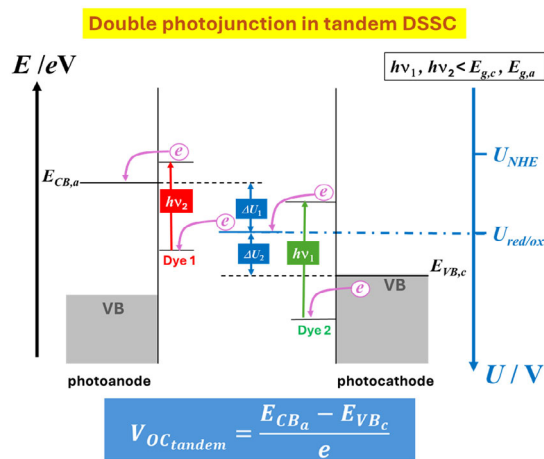


FIGURE 14 | Scheme of the frontier energy levels in a tandem DSSC. On the left Y-axis is shown the energy E (in eV) of the electrons in the photoelectrodes, dye-sensitizers and the redox couple. On the right Y-axis is shown the electrical potential U (in Volt/V). In the scheme the meaning of the symbols are $E_{CB,a/c}$ = lower energy level of the conduction band of the anode/cathode; $E_{VB,a/c}$ = upper energy level of the valence band of the anode/cathode; $E_{g,a/c}$ = energy bandgap of the anode/cathode; Dye1/2 = dye-sensitizer of the anode/cathode; $V_{OC_{tandem}} = \Delta U_1 + \Delta U_2$ = open circuit voltage of the tandem DSSC (in tandem DSSCs such a value does not depend on the oxidation potential value $U_{red/ox}$ of the redox couple); $h\nu_{1/2}$ = HOMO-LUMO energy separation for the dye-sensitizer 1/2.

of plants (traditional finality) and of electrical energy (finality of modern conception) [302]. The visible light transparency of the DSSC (or any other photovoltaic system) when the PV system is implemented in a greenhouse skeleton should warrant the transmission of the light necessary to the crop for realizing the chlorophyll photosynthesis while providing electrical energy for the most disparate demands (powering of sensors, automation of irrigation and water extraction from walls, powering of storage systems, etc.). The electrical energy provided by the visible-light transparent DSSC would be generated upon photoconversion of the solar photons in UV and IR ranges. This latter issue has motivated the search in the field of dye-sensitizers with high absorption in the NIR and near UV while abating visible light absorption (vide supra). Favorable consequences of the implementation of visible light transparent DSSCs for greenhouses or crop/land shielding span from the increase of land productivity or diminution of risks of agricultural product degradation to the savings of energy and water or to the diminution of the use of pesticides with resulting overall increase of sustainability in the agricultural practices [303, 304].

6 | Performance Under Indoor and Low-Light Conditions

6.1 | Photophysical and Electrochemical Determinants of DSSC Superiority Under Ambient Radiation

Artificial illumination delivers only a few tens of $\mu\text{W cm}^{-2}$ of predominantly visible photons, three to four orders of magnitude lower than terrestrial sunlight and essentially devoid of near-IR and UV quanta [305–308]. Conventional single-junction photovoltaics, whose junction energetics and carrier dynamics were optimized for 1 sun and for AM 1.5 G black-body illumination, suffer a triple penalty indoors: (i) spectral mismatch, (ii) reduced quasi-Fermi-level splitting, and (iii) intensity-dependent shunt and recombination losses. The detailed balance analysis of silicon under a CIE-B4 LED spectrum is illustrative: the theoretical limit collapses from 29.7% at 1 sun to 24.5% at 250 lx, with J_{SC} falling super-linearly owing to poor visible absorption and Auger-free radiative losses, while GaAs and CdTe, despite direct gaps, deliver only 40%–44% limit efficiencies under 1000 lx, all at the price of cost and toxicity. Detailed balance calculations by Ho and coworkers place the thermodynamic efficiency limit for a single junction operating under the CIE-B4 LED spectrum at 51%–57% when the absorber bandgap lies between 1.82 and 1.96 eV, with a striking 72% limit for monochromatic 555 nm radiation [309–311].

DSSCs are not encumbered by an intrinsic junction [312, 313]. Photon absorption, charge separation, and charge transport

are carried by three chemically orthogonal actors: molecular dye, mesoporous oxide, and redox mediator or hole transport material. Under low irradiance, the TiO_2 quasi-Fermi level ascends logarithmically ($\approx 59 \text{ mV dec}^{-1}$), while the injection efficiency of most metal-free push-pull dyes remains $>95\%$. The result is a much slower-than-expected increase of V_{OC} as the intensity drops from 100 to 0.5 mW cm^{-2} , provided that the mediator reduction potential is sufficiently positive and interfacial recombination is retarded. Freitag's copper-bipyridyl electrolytes, $[\text{Cu}(\text{tmby})_2]^{2+/+}$, push the equilibrium dye regeneration potential to $\approx 0.87 \text{ V}$ versus NHE yet impose only 100 mV of driving force; consequently, V_{OC} values of 1.10 V are routinely measured at 1000 lx with cosensitized XY1b/Y123 photoanodes [11, 15, 314]. In contrast, Γ^-/I_3^- requires $>500 \text{ mV}$ overpotential and caps V_{OC} at $\approx 0.75 \text{ V}$ [315, 316].

Charge collection is equally benign at low light. The logarithmic increase of the transport time constant τ_{tr} ($\tau_{\text{tr}} \propto I^{-0.35}$) is matched by an identical scaling of the recombination lifetime τ_{rec} ; thus, the collection efficiency $\eta_{\text{CC}} = 1/(1 + \tau_{\text{tr}}/\tau_{\text{rec}})$ remains >0.9 down to 50 lx. Parsons' impedance-normalized DLP (=digital light processing) light simulator demonstrates that a class-A spectral coincidence (0.91) and $<2\%$ spatial variance are now achievable, guaranteeing laboratory-to-field reproducibility (Table 1) [311].

6.2 | Molecular and Interfacial Engineering of Indoor-Optimized Dye/Electrolyte Couples

The spectral power density of a CIE-B4 LED peaks at 450 nm and has a Gaussian shoulder centered at 555 nm; FL-10 fluorescents add mercury-phosphor lines at 436 and 546 nm. Maximizing the absorbed photon-to-electron yield therefore demands dyes with extinction coefficients $\epsilon \geq 6 \times 10^4 \text{ M}^{-1} \text{ cm}^{-1}$ over 420–650 nm, negligible sub-400 nm absorption, and small Stokes losses. Such an emission is chemically accessible with triarylamine-thieno[3,4-*b*]pyrazine push-pull dyes, porphyrin-benzothiadiazole hybrids, or N-annulated perylenes; importantly, these chromophores need only span the 400–700 nm corridor that dominates artificial light. Figure 15 shows CIE LED-B4 and FL-10 lamp spectra [317] together with the absorption spectra of SL9 [318] and XY1b [319] and illustrates why DSSCs show unprecedented quantum yields under indoor conditions [7, 320, 321]. In the blue, at 450–470 nm, the dye extinction coefficient exceeds $1 \times 10^5 \text{ M}^{-1} \text{ cm}^{-1}$, while in the green–orange plateau (530–620 nm), it remains above $6 \times 10^4 \text{ M}^{-1} \text{ cm}^{-1}$; simultaneously, the copper(I/II) bis-bipyridyl mediator is optically silent across the entire range, thus eliminating parasitic absorption [312, 318, 319].

Cosensitization, once pursued mainly to panchromatically broaden outdoor DSSCs, has become an indoor design lever for suppressing intersensitizer quenching and sculpting an absorption

TABLE 1 | Specifications for light source classification for indoor PV (IPV) measurement [311].

Class	SA	A	B	C
Spectral coincidence	0.95–1.05	0.75–1.25	0.6–1.4	0.4–2.0
Spatial nonuniformity	within $\pm 2\%$	within $\pm 2\%$	within $\pm 5\%$	within $\pm 10\%$
Temporal stability	within $\pm 0.5\%$	within $\pm 0.5\%$	within $\pm 2\%$	within $\pm 10\%$

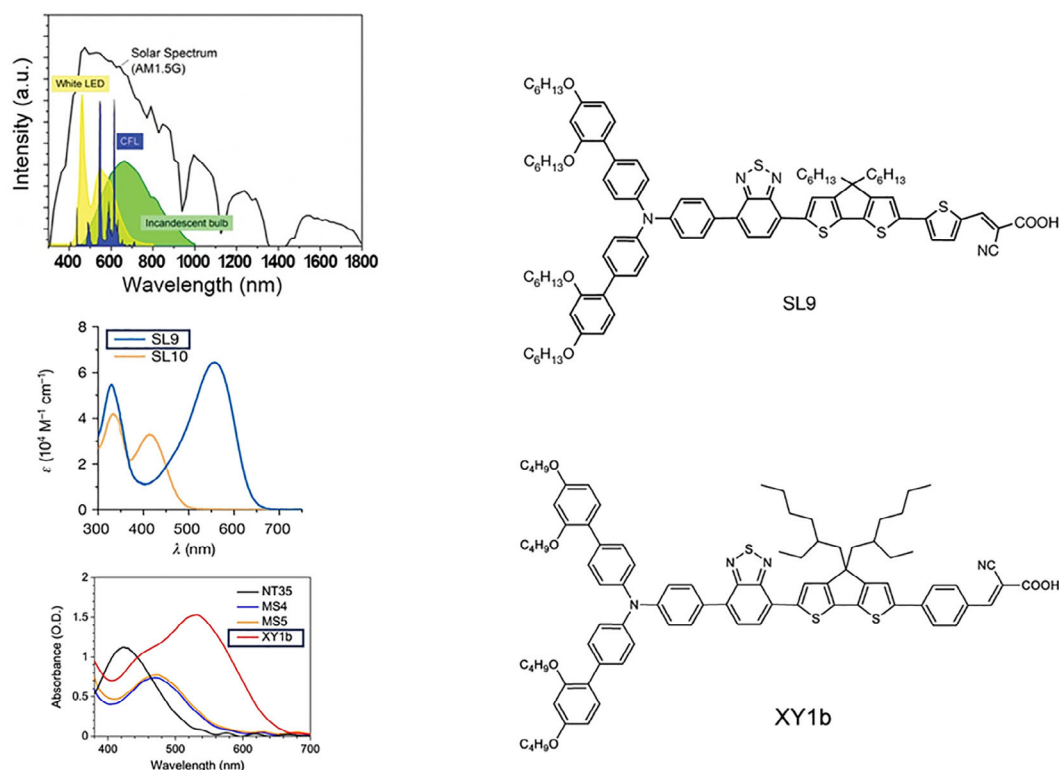


FIGURE 15 | Left column: (top) spectra of the solar light (black trace), white LED (yellow-shaded profile), compact fluorescent lamp (CFL, blue-shaded profile), and incandescent bulb (green-shaded profile)—reproduced with permission from ref. [317]. Copyright 2021, Wiley; optical absorption spectrum of (middle) SL10 – reproduced with permission from ref. [318]. Copyright 2023, Springer Nature Publishing Group – and (bottom) XY1b dye-sensitizers – reproduced with permission from ref. [319]. Copyright 2021, Springer Nature Publishing Group. Right column: molecular structures of (top) SL9 and (bottom) ZXY1b dye-sensitizers.

envelope that mirrors LED or fluorescent emission. The classic SL9/SL10 dyad of Grätzel already illustrated how complementary spectra and facet-selective binding [SL9 on rutile (100) and SL10 on anatase (101)] [322] elevate low-lux fill factors to 30% PCE at 1000 lx. Building on that principle, Michaels et al. Coadsorbed the blue-biased azabenz-annulated LEG1 dye with the broadband push-pull chromophore XY1 on TiO₂, forming a dense, iso-absorptive monolayer that flattened the IPCE to unity across 420–680 nm; paired with a [Cu(tmby)₂]^{2+/+} electrolyte, the LEG1||XY1 architecture delivered a record 38% PCE and 1.0 V *V*_{oc} under 1000 lx fluorescent light, enabling self-aware IoT nodes powered solely by ambient energy [6, 318, 323, 324]. Future gains are expected to arise from choreographing multidye films whose collective extinction remains constant out to 720 nm.

Electrolyte design has lagged behind dye chemistry until recently. Copper and cobalt complexes now offer a synthetic phase space spanning 0.70–1.00 V versus NHE with ligand-controlled reorganization energies $\lambda < 0.3$ eV. Continuous-flow exchange experiments show that [Cu(tmby)₂]^{2+/+} withstands more than 10⁶ redox cycles without ligand scrambling, provided that 4,4'-disubstitution avoids pyridine dissociation. The remaining bottleneck is diffusion: the DNP NMR-measured Dox of 6×10^{-6} cm² s⁻¹ in MeCN halves in protic ether cosolvents used for viscosity suppression. Molecular engineering of zwitter-ionic spacers (MS5) yields larger hydrodynamic radii but doubles the cation-independent self-exchange rate k_{ex} to 1.8×10^8 M⁻¹ s⁻¹, enabling $J_{\text{SC}} > 160$ $\mu\text{A cm}^{-2}$ at 200 lx [318].

In Table 2 are listed the features of performance for DSSCs and PVs devices assembled for use in indoor conditions when illuminance is 1000 lx.

Surface passivation remains a decisive variable in indoors because the quasi-Fermi level is close to the conduction band edge. Preadsorbed hydroxamic acids elevate the Helmholtz potential, shifting EC by –60 mV and reducing the recombination resistance with Cu^{II} by a factor of four. Similarly, Al₂O₃ atomic-layer over-coatings of 0.7 nm cut the interfacial rate constant $k_{\text{CB-Cu(II)}}$ from 45 to 2 cm s⁻¹ without perturbing the value of the injection time, τ_{inj} [200].

6.3 | Industrial Outlook: Self-Powered IoT, Standardization Ad Market Trajectory

IoT is scaling toward 125 billion nodes by 2035, with aggregate electricity demand forecast to top 1 PWh a⁻¹ [325]. Office and retail lighting operates in the illuminance range of 500–1 000 lx for 3 000 h year⁻¹; at 150 $\mu\text{W cm}^{-2}$ (500 lx), a 2 cm² DSSC can sustain a bluetooth low energy (BLE) mesh sensor at 10 μW average draw, displacing coin cells whose embodied energy is 25 kJ (vs. 1 kJ for the DSSC module) and eliminating 7 kt year⁻¹ of Li battery waste in the UK alone. IEC TS 62607-7-2:2023 establishes the Indoor Standard Testing Conditions (50–200–1000 lx; CIE B4, FL-10 spectra; 25 °C), enabling bankable performance metrics. DLP-based light simulators meeting class-A spectral coincidence

TABLE 2 | Performance of Leading Indoor DSSCs and inorganic PVs at 1000 lx.

Technology	V_{OC} , mV	J_{SC} , mA cm ⁻²	FF	PCE, %	Notes/device details
DSSC [10]	870	20.1	0.75	28.9	Co-sensitized XY1b/Y123; electrolyte: [Cu(tmbpy ₂)] ^{2+/+} ; area: 2.8 cm ² ; lamp: CIE FL-1
DSSC [275]	878	149.3	0.77	31.8	Co-sensitized XY1/Y123; electrolyte: [Cu(tmbpy ₂)] ^{2+/+} ; area: 2.8 cm ² ; lamp: OSRAM Warm White 930
DSSC [6]	910	36.7	0.77	34.0	Co-sensitized L1/XY1; electrolyte: [Cu(tmbpy ₂)] ^{2+/+} ; area: 2.8 cm ² ; lamp: OSRAM Warm White 930
DSSC [244]	995	147	0.78	38.0	Co-sensitized L1/XY1; electrolyte: 0.06M [Cu(tmbpy ₂)] ^{2+/+} ; area: 0.196 cm ² ; lamp: OSRAM Warm White 930; machine learning drive
DSSC [282]	980	138	0.81	34.5	Co-sensitized MS5/XY1b; electrolyte: [Cu(tmbpy ₂)] ^{2+/+} ; area: 2.8 cm ² ; lamp: OSRAM Warm White 930
Polycrystalline-Si (pc-Si) [271]	250	81.2	0.48	6.1	Standard c-Si of commercial type; lamp: CIE FL-1; area: 1.68 cm ²
Amorphous-Si (a-Si) [271]	2690	44.8	0.56	10.9	Standard a-Si of commercial type; lamp: CIE FL-1; area: 3.18 cm ² ; four cells in series
GaAs [275]	940	99	0.80	19.0	Single junction cell; lamp: CIE FL-1

(0.91), $\pm 2\%$ uniformity, and $\pm 0.5\%$ temporal stability across three decades of illuminance [281, 326–329].

On the manufacturing side, several suppliers have reached pilot-line throughput [330]:

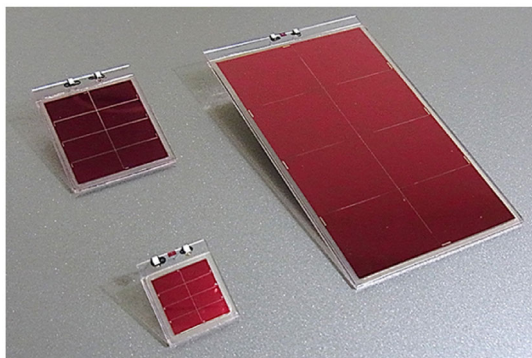
1. Exeger (SE): Roll-to-roll slot-die deposition on 1 μ m ITO-PET with Cu(tmbpy)₂ electrolyte and PEDOT current-collection grid. Mass-market integration started in 2022 in audio headsets (Urbanista Los Angeles), yielding 240 μ W cm⁻² at 1000 lx with zero user charging.
2. Ricoh (JP): Fully solid-state “EH-DSC” employing polyimide-gelified Cu-phenanthroline redox shuttle, laminated into wireless IoT tags. Life test: 93% PCE retention after 1000 h at 45 °C under 1000 lx (Figure 16).
3. Fujikura (JP): Glass-on-ITO Cu-complex modules powering BLE multisensor beacons, exhibiting 50 μ W cm⁻² at 500 lx, and certified for –20 to 80 °C.
4. Ambient Photonics (US): Printed DSSC-based modules optimized for indoor ambient light, delivering up to ~ 150 μ W cm⁻² at 1000 lx fluorescent light, designed to power

wireless IoT devices and sensors in commercial and residential environments, with proven operation across the temperature range of 0–50 °C.

Bill of materials teardown indicates a present module cost of 6–8 USD m⁻² at a 1000 m² a⁻¹ scale, dominated by dye (43%) and patterned FTO (31%). Transitioning from screen-print sintering to photonic flash-anneal TiO₂ and from Ru-free metal-free dyes to gram-scale Y-series indolorethene donors cuts capex and halves embodied CO₂ relative to Li-ion coin cells.

To consider is that 8 times more energy can be saved by autonomous IPV than is consumed in their manufacture (compressed.pdf slide on IoT energy). The addressable indoor-PV market is projected to exceed 1 B USD in 2026 with 70% CAGR, dominated by environmental sensing (33%), asset-tracking (29%), and smart home (18%). Remaining challenges are convergence level, electrolyte water-vapor diffusion through ultrathin encapsulants, and operational stability under pulse width modulation-driven LED spectra which contain 25% UV-violet content. Multiwavelength stress tests show devices lose 4% PCE after 1000 h at 0.2 sun equivalent when Al₂O₃/SiO₂ nanolaminate barriers are applied, hinting at 10-year indoor durability. Color-rendered semitransparent modules for BIPV remain a secondary but growing niche. H-Glass façade at Science Tower (Graz) delivers 12 W m⁻² under daylight with 500 m² of red-filter DSSC; ongoing retrofit projects specify Cu-electrolyte devices to avoid electrophoretic iodine degradation seen in Lausanne modules (Visual 5: compressed.pdf slide) [311, 323, 331].

Indoor photocapacitors present a compelling solution where maintenance-free electronics require burst power beyond what photovoltaics alone can supply. Recent advances demonstrate that integrated dye-sensitized photocapacitors (PCs), or energy-storing DSSCs (ES-DSSCs), achieve seamless indoor light harvesting and storage by coupling a DSSC photoanode with an asymmetric polyviologen-based supercapacitor, unified by a shared PEDOT CE [332]. Such three-terminal hybrids exploit

**FIGURE 16** | Indoor DSSCs models by Ricoh.

quasi-solid-state copper complex electrolytes and bio-derived fungal membranes to suppress leakage and recombination, delivering photocharging voltages up to 920 mV and overall efficiencies up to 18% under 1000 lux ambient light. This design retains 95% capacity after 1000 h of operation, sustaining low-power edge AI tasks and wireless IoT networks with 3.5× higher inference throughput than amorphous silicon modules. Viologen-based electrodes with V-shaped PVN architectures minimize reorganization energy (0.1–0.2 eV), enhance cycling stability beyond 3000 cycles, and mitigate self-discharge via suppressed Cu^+ crossover. These findings establish integrated photocapacitors as the natural convergence point for indoor PV, energy storage, and power-aware AI microcontrollers, advancing maintenance-free, autonomous IoT deployments [333–335].

Soon, three converging vectors will propel indoor DSSCs beyond 40% PCE: (i) halogen-bonded auxochromes and mixed siloxy/hydroxamate anchors pushing dye extinction to 720 nm while retaining $E_g \approx 1.8$ eV; (ii) tetradentate copper cages with $\lambda < 0.15$ eV and $k_{\text{ex}} > 2 \times 10^8 \text{ M}^{-1} \text{ s}^{-1}$, enabling $V_{\text{oc}} \geq 1.15$ V in a single junction; and (iii) multilayer $\text{Al}_2\text{O}_3/\text{TiO}_2$ encapsulation with water vapor transmission rate $< 5 \times 10^{-4} \text{ g m}^{-2} \text{ d}^{-1}$, projecting ten-year indoor durability. Parallel advances in edge AI (sub-10 μJ inference) will couple seamlessly with harvester-aware firmware, delivering a battery-less, self-aware ambient internet.

7 | Manufacturing and Scalability of DSSCs

As the global energy transition accelerates, the scalability and manufacturability of PV technologies have become critical criteria in order to widespread their distribution. Amongst next-generation PV solutions, DSSC technology stands out due to its compatibility with low energy consumption and scalable fabrication methods. In particular, 3rd generation of PV technologies uses printing methods that are already well-developed techniques in other industry sector such as paper and textile industry, as well as silicon-based PVs to cover part of its production process. In fact, it is with the advent of 3rd generation PVs that printing/coating techniques (i.e., screen and pad printing, blade, spin, and spray coating, slot-die deposition) have become widely used across nearly all the fabrication processes [336]. Such coating techniques can be carried out at different temperatures depending on rigidity of the substrate: if flexible below 180 °C and if rigid not above 500 °C.

As already anticipated, DSSCs are photoelectrochemical devices that possess all the characteristics ideal for serving in emerging PV applications such as BIPV, indoor energy harvesting for IoT devices, and agrivoltaics [76, 337]. Their intrinsic ability to perform efficiently under diffuse and low-light conditions—enabled by their mesoporous architecture that supports omni-directional light harvesting—imparts DSSC technology a particular behavior that sets it apart from traditional semiconductor-based photovoltaics. In fact, DSSCs exhibit a nearly constant PCE regardless of their orientation to incident light and are inherently bifacial [7], thanks to their transparent counter-electrodes. These characteristics make them particularly well-suited for architectural integration, where light can enter from multiple directions and

surfaces. Within the broader category of transparent photovoltaic technologies, DSSCs are notable for offering high visible transparency (typically over 50%) and excellent solar radiation transmission, often exceeding 60%. In BIPV contexts, two optical parameters are especially critical: the AVT, which measures the fraction of visible light (380–780 nm) that passes through the material (vide supra) and CRI that assesses how accurately colors are perceived under the transmitted light compared to natural sunlight. DSSCs offer precise tunability of both AVT and CRI by varying the dye composition and adjusting the photo-electrode thickness. Remarkably, DSSCs have demonstrated AVT values up to 78% and CRI values as high as 92.1% [42], underscoring their potential not only for efficient energy generation but also for an aesthetically effective integration into modern building designs.

7.1 | Roll-to-Roll, Screen Printing, and Doctor Blade Methods

To lead the energy transition, it is essential that the DSSC technology passes from lab-scale research to industrial-scale production, adopting scalable, high-throughput, cost-effective manufacturing methods and low energy consumption technologies with respect to traditional PV technologies such as the silicon-based ones. Among these, roll-to-roll (R2R) processing stands out as a viable approach, and it enables the fabrication of DSSCs both on rigid and flexible substrates using automated process since DSSCs are less prone to contamination with respect to PV technologies that require high-purity silicon and O_2 -free environment of processing/assembly and can be processable at ambient temperature [338]. In general, R2R is especially suited for manufacturing lightweight, portable, and wearable—a possible field of application for DSSCs—solar devices due to its compatibility with low-temperature processes and plastic materials.

Screen printing is the most widely adopted method for DSSC production, facilitating the deposition of key functional layers—such as the TiO_2 photo-electrode and the CE—with an elevated precision in thickness and patterning. Its scalability makes it ideal for industrial manufacturing; however, large-scale uniformity and reproducibility require tight control over printing parameters and ink formulations [336].

The doctor blade system is also an effective technique for depositing solution-processable layers such as TiO_2 [339]. Compared to screen printing, doctor blade technique offers simpler operation and is particularly useful when working with low-viscosity pastes or inks. Its ability to produce uniform, reproducible coatings make it an effective bridge between lab-scale methods and full-scale production.

7.2 | Challenges in Large-Area Uniformity and Reproducibility

Despite the availability of scalable fabrication techniques, several technical challenges arise when DSSCs need to be manufactured over large surface areas (larger than an A4 sheet). One of the primary concerns is achieving uniform film deposition across an entire dye-sensitized solar module (DSSM). For example,

variations in TiO_2 layer thickness or dye loading can lead to inefficient photo-response and reduced device PCE. Ensuring the distribution of the electrolyte species is also challenging; in fact, air gaps or incomplete filling can compromise the electrical performances and long-term stability [340, 341]. Sealing and encapsulation become increasingly critical at larger scales. For example, optimal sintering of the photo-electrode occurs at above 500 °C. After the sintering process in a furnace belt (the one suitable for large-scale production), there is an evident deformation in the Z-axis (around 300 μm from the center of the substrate to its edges) [342] of a SLG (soda lime glass)/FTO (fluorine doped tin oxide) the area of which is hundred cm^2 substrate. This occurrence reflects in an inadequate sealing and can result in electrolyte evaporation or moisture ingress, accelerating degradation. Furthermore, as module dimensions increase, interconnect and current-collection design becomes the primary aspect of FF and energy yield: stripe geometry and layout must be optimized together with TCO sheet resistance, finger/busbar resistivity, and interconnect/contact resistances to limit voltage drops and series resistance losses while preserving transparency and aperture ratio [343–347].

In general, at the module level, the factors of sheet resistance, interconnect design, series resistance, nature of the electrolyte, and relative management in terms of containment as well as the strategies of cell sealing, especially in the case of fluid electrolytes, dictate directly the values of overall power output and efficiency through the variation of the resulting fill factor of the DSSC. It is proved that a too high series resistance in the DSSC flattens the slope in the proximity of the open-circuit voltage with consequent diminution of the fill factor and overall conversion efficiency also in DSSCs similar to what occurs in junction-based photovoltaic technologies. Analogously, in DSSC (photoelectrochemical devices in which both electronic and ionic transport takes place), the resistive terms determine the internal power losses I^2R (I being the total electrical current passing through the cell and R the total resistance of the cell) and affect the thermal stress due to the heating caused by the Joule effect. Therefore, the choice of TCO with low sheet resistance ($<10 \Omega/\square$) [348] and electrolytes with high electrical conductivity (a compositional term) and large electrical conductance (a geometrical/configurational term) is of utmost importance. In turn, the sealing procedure with its efficacy of confinement of the fluid components affects primarily the degradation of the cell under operative conditions [349]. The incorrect management of the electrolyte in wet photovoltaics, as the case of the DSSC technology, incurs in liquid electrolyte leakage for spilling or evaporation with irreversible dry-out and fatal, irreversible failure of the photoelectrochemical cell. Because of that, it appears quite evident that self-standing and self-contained solid-state/gel-like electrolytes have to be generally preferred provided that the charge/mass transport properties of both electrolytic and redox components in the solid-state/gel-like electrolytes are sufficiently good to sustain an appreciable electrical current. At this concern it is important to achieve solid/gel electrolytes with not too high electrical resistance, being the latter a term that constitutes the series resistance for a PV and DSSC system. If the DSSC sealing lacks integrity, the defective DSSC can present a relative diminution of the short-circuit current with respect to a well-sealed DSSC due to the increase of the resistance associated to mass transport and charge transfer through the electrolyte for the

permeation of moisture and oxygen (a redox species), loss of redox mediator, and acceleration of degradation reactions between metal oxides and water (dissolution), contact metals and water/oxygen (corrosion), with resulting delamination and collapse of the layout. With the increase of the photoactive area in DSSCs augments the risk of nonuniformity of the current flow due to possible nonuniformity of the chemical-physical properties of the electrically conductive layers along the surface and the nonhomogeneity of the electrically conductive layers across the thickness. Moreover, effects of transverse electrical resistance associated with charge transport parallel to the surface of the electrically conductive layers can occur if the geometry of contacting with the external load for charge collection is strongly asymmetric with respect to the planar configuration of the conductive layers. All these latter factors are related somehow to the anisotropic features of the electrically conductive materials and the bidimensional geometry of the system and can lead to the creation of hot spots that accelerate thermal degradation (current crowding effect) and possible local modification of the electrical conductivity and mechanical properties. The eventual presence of sharp corners, cracks, voids, bends, and right angles (especially in cell configurations with deliberately nonplanar configuration as in the case of DSSCs onto flexible plastic substrates) forces the electric field lines to bunch together and brings about the spiking of the local current density. At the engineering level, for the preparation of multi-DSSCs modules, the use of rounded corners, multifinger geometries, interdigitated layouts (without affecting severely the optical transmission properties of the DSSC), multi-busbar, shingled designs with adhesive conductors, and auxiliary series strings is preferred for the prevention of the nonuniformity of the electric field lines, for smoothing out the paths of electrical current, and the reduction of edge concentrations. Such an engineering approach is favorable also in terms of mitigation of the potential-induced degradation [269, 350].

As a conclusive remark of this section, establishing robust and efficient interconnections between individual cells becomes more complex and must be carefully engineered to prevent performance loss. Reproducibility is another crucial issue in large-scale DSSC manufacturing. Minor variations from one substrate to the next one in material properties—such as a not proper dye adsorption or a not uniformity in thickness of the deposited layers can significantly affect device' performances. Thus, the standardized and closely monitored production protocols are crucial to ensure a robust quality that enables the commercial viability of DSSCs.

7.3 | Cost-Effective Materials and Fabrication

Manufacturing routes based on low-temperature processing and rapid curing/annealing enable low-thermal-input DSSC fabrication by reducing the overall thermal budget of device fabrication [338, 351, 352].

As previously outlined, one of the defining strengths of DSSCs is their potential energy-efficient fabrication. The key materials involved, such as titanium dioxide, are abundant, nontoxic, and relatively cheap. From the typical costly ruthenium-based dyes, the research has moved to toward metal-free organic dyes, porphyrins, which offer cost and sustainability advantages without significantly compromising performance [353–361].

Alternatives to platinum CEs, such as carbon-based materials (e.g., graphite, carbon black, or graphene) [343], can reduce costs while maintaining acceptable efficiencies. Overall, the combination of scalable and low-energy consumption processes and adaptable form are factors that render DSSCs an attractive PV technology. While DSSCs may not yet match silicon in peak efficiency, their advantages in terms of aesthetics, flexibility, diffuse light performance, and manufacturing simplicity make them ideal for emerging solar applications where these factors are prioritized.

Bringing DSSC technology from the laboratory scale to the large-scale production involves a multifaceted approach that faces several challenges. First of all, the technical steps need to be addressed, such as optimization of materials, performance improvement, and scaling-up of both the devices and the production. Moreover, also managing aspects must be taken into account, such as marketing analysis and strategy development. Amongst these challenges, the effective capability of scaling-up the DSSCs while ensuring a high reproducibility, is a mandatory and crucial step toward the industrialization. Indeed, the PCE's record (15.2%) has been achieved on an active area equal to 0.12 cm² active area [318], while a PCEs equal to 8.8%–10.7% have been reported by Sharp for a DSSM of 400 and 27 cm² of active area, respectively [362]. A DSSM is the unit of the scaling-up process since it represents the joining link between a single cell (whose active area ranging from less than 0.25 to 5 cm²). In a DSSM, the active area (AA) is designated for solar energy conversion, while the dead area (DA) accommodates interconnections and provides their necessary protection. The aperture ratio is defined as $AR = AA / (AA + DA)$, where the sum of AA and DA is the aperture area (APA).

The AR (also known as geometrical fill factor, gFF) is one of the key parameters for BIPV application since it determines the power generated by the effective area occupied by a solar module or panel, that is, the efficiency calculated on APA takes into account the areas that does not contribute to photocurrent generation. BIPV deployment requires explicit module-level engineering. In large-area DSSMs, performance and yield are strongly affected by (i) Ohmic losses driven by TCO sheet resistance and lateral current collection, (ii) additional series-resistance contributions from busbars and contact resistances, (iii) geometric losses due to dead area needed for scribing and interconnect protection (AR), and (iv) electrolyte management (bubble-free filling and uniform distribution) together with strong sealing to prevent evaporation and moisture ingress under outdoor thermal cycling [41, 343–346].

A DSSM requires a specific design to overcome the electrical limitation due to the sheet resistance of the TCO (Transparent Conductive Oxide), that is, the FTO [363]. At the module level, the voltage drop along the TCO can become comparable to the operating voltage per stripe, directly lowering the FF; therefore, the module must be designed by adopting an explicit series resistance “ensemble” that includes TCO lateral resistance, finger/busbar resistance, and interconnect/contact resistances. In Z-type DSSMs, the vertical interconnect material and its protection against electrolyte corrosion are likewise critical to preserve long-term series connection integrity in BIPV operation [343, 344, 346]. The cells of a DSSM can be connected in parallel

(sum of the currents) or in series (sum of the voltages). Four possible modules' configurations can be realized: parallel, series monolithic, series W-type (where the polarity of adjacent cells is inverted), and series Z-type (where adjacent cells present the same polarity) [343]. The abovementioned efficiency record on DSSMs have been achieved in series configuration. In the Z-type configuration, the module is composed of identical cells sandwiched between two TCO-covered glass, with conducting vertical interconnections (typically made of silver) connecting adjacent cells. Basically, the sequence of materials for an electrode of a Z-type DSSM is scribe/silver/TiO₂ (if photoactive electrode, PE) or Pt (if CE, CE), and so on. Making effective the designed distances between each element for every device fabricated is pivotal both for the realization of the wanted aperture area of one cell within a DSSM and for its repeatability within a single DSSM (Figure 17, vide infra). Since DSSMs can be fabricated by adopting all main printing techniques, the requirements to address the desired layout can be fulfilled—for example, via a screen-printing deposition of the materials involved [337].

Achieving an accuracy of the order of tens of micrometers (within X–Y plane) in the layout of a DSSM is a fundamental aspect that enables the creation of complex devices with maximized active area and an overall enhanced transparency. Only automated manufacturing processes can guarantee the necessary repeatability and precision to advance DSSC technology toward broader commercial applications.

As depicted in Figure 17, the value of the interdistance between two adjacent cells determines the dead area [364]. Minimizing the dead area is the main objective within the realization of a DSSM and, in general, for a solar device. However, a minimum DA is typically unavoidable because it must accommodate scribing lines, interconnection regions, and insulating/sealing regions that isolate metallic fingers (e.g., Ag) from the electrolyte to prevent corrosion and shunts; hence, DA reduction must be balanced against interconnection reliability for BIPV modules [343, 344, 346]. The interdistance between two adjacent cells is set by an already designed screen (i.e., the main toll of the screen printing deposition technique), and an accuracy of tens of micrometers can be achieved only by employing an automated, camera-controlled process.

While the AR (or geometrical fill factor, gFF) quantifies the fraction of the module area that is optically active (i.e., exposed to light), the scale-up of DSSMs is limited by electrical losses that grow with the lateral collection length (“size effect”). The series resistance increases with device width because photocurrent must travel laterally through the TCO before reaching the busbars, leading to a progressive FF penalty if the layout is not optimized. This aspect is well explained for DSSC scale-up in terms of substrate/TCO resistance and module geometry, highlighting the need for optimized current-collection schemes at large area [363, 365].

Accordingly, the design variables in Figure 17 (finger width, interconnection distance, and cell width) should be interpreted as a trade-off between (i) minimizing dead area and (ii) mitigating resistive losses. Beyond the TCO sheet resistance, contact resistance at metal/TCO and at vertical interconnect interfaces becomes increasingly relevant when contact areas are reduced

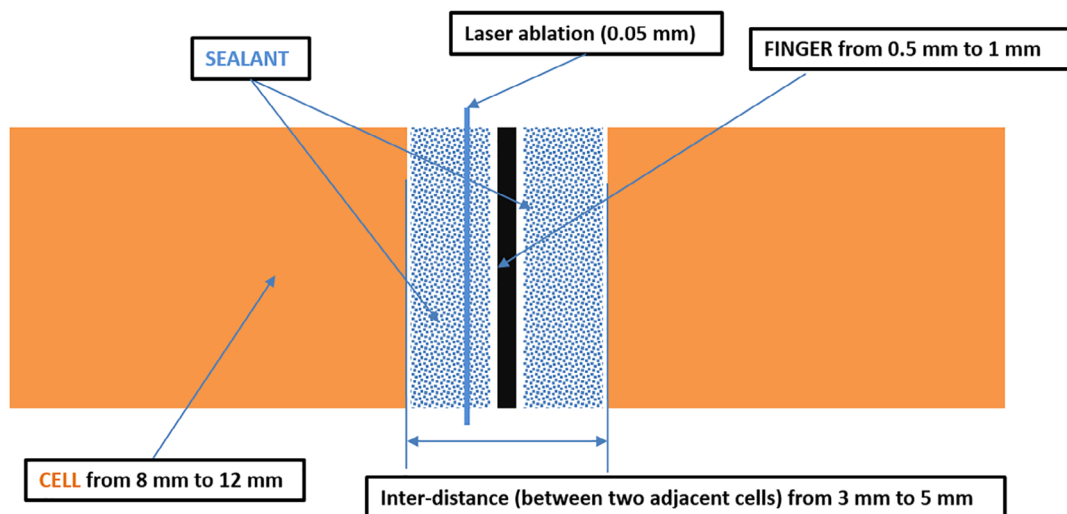


FIGURE 17 | Ranging width of a cell, a finger and of a distance between two adjacent cells within a DSSM. The optimal width for a DSSM' cell ranging from 8 to 12 mm [317]. In orange the AA, the whole area between the two cells is the DA. Its width is the fundamental parameter for scaling up DSSC technology.

or when alignment tolerances lead to nonideal overlap regions, directly contributing to the effective series resistance and FF losses. Introducing engineered current collectors (e.g., sputtered metal lines protected by conductive coatings, embedded collectors, or corrosion-resistant interconnect schemes) can substantially improve current collection and FF at module scale [366].

The higher the possibility of reducing the gap between two adjacent cells and managing the thickness of the fingers (typically ranging from 15 to 25 μm), the higher the power generated, as this both increases the active area and optimizes the distance between CE and PE with an accuracy of tens of micrometer in $X-Y$ plane and less than 10 μm along the Z -axis [367].

In Figure 18 the structural sequence of a DSSM's layout is described. The initial laser scribing step (width of 0.05 mm) establishes the reference geometry for all subsequent layers. The process flow proceeds with the deposition of metallic (typically silver) fingers, followed by the application of the photoactive cell layers. Given the narrow intercell spacing (3 to 5 mm) and fine finger widths (0.5 to 1 mm), the cumulative effect of reducing misalignments can significantly decrease imperfections in the active areas, directly impacting module's performance, mainly the FF. Only with the capability to precisely control the full alignment chain—from the scribing through the finger

layout to the active layer definition—can one ensure a compact, high-performance design that maximizes the gFF. This level of accuracy is not merely a matter of aesthetics or manufacturability but is fundamentally tied to the module's power output and commercial viability. Regarding the latter point, the capability of properly modifying the printing parameters according to paste/ink formulations adopted in the printing process makes possible a strict control of the finger's thickness. Indeed, this latter parameter is crucial for a DSSM Z-type since it determines the amount of the sealant (typically a commercial UV-curable resin) needed for a specific value of thickness. Moreover, even the shape of the sealant is determined by the layout in $X-Y$ plane. After printing the electrodes, the PE is immersed in a dye solution, typically overnight. Then, PE and CE are sealed together and aligned in series connecting the fingers. In this regard, the automated process for the alignment of the PE' fingers and the CE' fingers for the vertical contact is mandatory, ensuring an accuracy of tens of micrometers in $X-Y$ plane. Finally, a thermoplastic (Bynel or Surlyn) gasket (made of a foil thick around 60 μm) is shaped according to the module layout and then placed between PE and CE to assembly the device. The stack formed by the two electrodes and the thermoplastic foil is inserted in a thermal press. It is melted at a pressure of around 2 bar and a temperature of around 150 $^{\circ}\text{C}$ for 1 min, becoming transparent after polymerization. An alternative method to assemble the two electrodes is

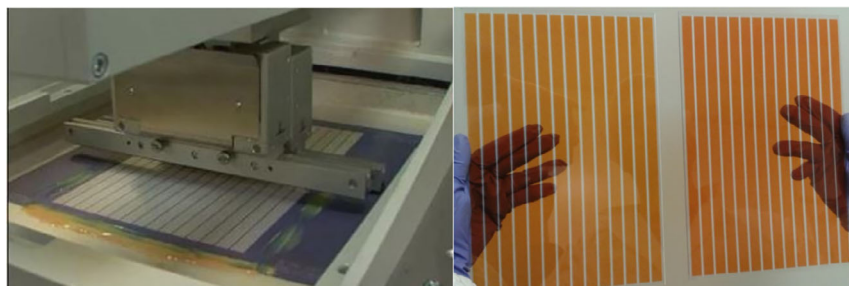


FIGURE 18 | Left: deposition—via screen printing technique—of a TiO_2 paste onto large area substrates (600 cm^2). Right: photo-electrodes after dye dipping. Different colors and transparency are shown [368].

depositing the sealant via dispensing according to the layout of the DSSM. In this case, the material is a commercial UV curable resin which has to be treated under UV radiation during the assembling step performed in an ad hoc press customized for the UV treatment. After the assembling step, an electrolyte (typically containing a triiodide/iodide redox couple) is injected in the DSSM by a vacuum back filling system through 60 μm slits (one per cell) shaped in the gasket [368] or drawn *via* dispensing. At the module level, electrolyte management must ensure complete, bubble-free filling of each cell stripe and stable wetting over large areas because trapped air or incomplete filling can translate into local series resistance increases and accelerated degradation. In BIPV, sealing must also guarantee a long-term hermetic sealing, not only at the perimeter but also at electrolyte injection holes, under outdoor humidity and thermal cycling. Besides polymeric sealants (thermoplastic gaskets or UV-curable resins), glass-frit encapsulation has been proposed as a route to improved hermetic sealing, although sealing the electrolyte injection holes remains a key manufacturing challenge [346, 347]. Thus, both the space near each vertical contact and the entire frame of the DSSM are filled with the sealant (thermoplastic or UV-curable resin), which has the role of both assembling the two electrodes and protecting the fingers (made of silver) from the corrosive behavior of the electrolyte. In this way, by employing advanced manufacturing techniques that ensure high layout accuracy, it is possible to create devices with larger active areas while simultaneously increasing the overall transparency. Automated manufacturing processes play a pivotal role in achieving the precision required in DSSC production. The reproducibility provided by automated systems ensures that once a successful design is established, it can be replicated accurately (within tens of micrometer in the X-Y plane and less than 10 μm in Z-axis) across multiple units. This consistency is crucial for scaling up production to meet industrial demands.

At the module scale, the device architecture, such as that shown in Figure 18, determines both lateral current collection through the current collectors (i.e., TCOs) as well as vertical charge collection across the various interfaces and interconnection areas, thereby strongly impacting series resistance and overall module performance.

Thus, parameters such as photoanode thickness and uniformity, device size (i.e., collection length), and contact area in the interconnection regions directly influence whether performance is limited by photogeneration or by collection losses. Reduced contact area, longer collection paths, or thickness nonuniformities can lead to increased series/contact resistance, nonuniform current extraction and FF losses, and, in extreme cases, local transport bottlenecks and enhanced recombination. Overall, these considerations underline the need to cooptimize geometry, interconnect/contact design, and process uniformity when scaling DSSCs to large-area DSSMs [369, 370].

Despite the extensive literature available on DSSCs, there are only few studies focused on DSSMs or panels (i.e., more DSSMs combined to form a larger panel). In the past decade, the most relevant works—in terms of PCE and future reliability (i.e., fabrication of panels or preparation of electrolytes with sustainable features [371–373] – are the following: In 2016, a large-area device (10 modules of 225 cm^2 , arranged in a panel) with an

initial PCE equal to 1.23% was reported [374]. In 2017, a 900 cm^2 DSSM for BIPV was reported [344], showing a PCE equal to 3.19%, while in ref. [375] the PCE value was not clearly reported. In 2018, [345], nine transparent DSSC sub-modules were assembled to form the unit size of a DSSC BIPV glazing of 0.975 $\times$ 0.965 m, showing an operating PCE (in the case of the vertical DSSC BIPV window) ranging from 2.65 to 4.14%, with an average of 3.40%. In 2021, a PCE of 3.9% on a 221 cm^2 active area was achieved, with a transparency in the module's aperture (312.9 cm^2) area of 35% [337], suitable for greenhouse application [346]. In fact, 40 of these DSSMs were assembled to form ten panels (each made of 4 DSSMs) to filter the light from the roof of the greenhouse [337]. The maximum PCE for a panel (in outdoor conditions) was equal to 3.83%.

Regarding flexible substrates, in 2016, modules developed on substrates of 100 cm^2 were reported in two different works [376, 377], with a PCE equal to ~3% to 3.27%, respectively. Regarding indoor environment, in 2018, a module with an active area of 26.8 cm^2 obtained an overall efficiency of 11.94% under 1000 Lx [378], outperforming its reference (in terms of dye) counterpart (PCE = 11.5%). In this work reporting the ~12% indoor DSSM, the device's PCE is not measured or reported under 1 sun (1 kW/m^2 , AM1.5G).

The disparity between small-area-based papers and module/panel-based papers highlights the need to address challenges and optimize processes at the module level, where scalability and durability play a critical role.

To bridge this gap, it is essential to explore alternative approaches, such as employing pilot fabrication lines. A pilot line equipped with highly automated machines enables the investigation and refinement of fabrication processes under conditions closer to industrial manufacturing, allowing for insights that cannot be fully captured in small-scale laboratory settings, such as device's reproducibility and a deeper comprehension of bottlenecks during the manufacturing steps. At present, in literature, a PCE of 5.6% has been reported on a Z-type DSSM with an AA of 600 cm^2 (gFF around 80%) device realized in a pilot line facility [76].

8 | Comparative Positioning of DSSCs Within the Photovoltaic Landscape

To place DSSCs within the wider photovoltaic landscape, Table 3 compares them with organic solar cells, single-junction perovskite solar cells, monolithic perovskite/silicon tandem devices, and commercial crystalline silicon technologies [73, 144, 318, 368, 376, 377, 379–410]. The table provides a comparative analysis of the different technologies based on different indicators, from PCE under one sun and indoor illumination to operational stability and recyclability. Stability and life cycle data should also be interpreted with care as device area, encapsulation strategy, aging protocol, assumed lifetime, functional unit, and end-of-life scenario vary considerably across the literature.

Overall, the comparison does not suggest that DSSCs outperform the other photovoltaic technologies in terms of PCE peak,

TABLE 3 | Comparison of dye-sensitized solar cells (DSSCs), single-junction halide-perovskite solar cells (PSCs), monolithic two-terminal perovskite/c-Si tandems, commercial c-Si photovoltaics, and organic solar cells (OSCs).

Category	Dye-sensitized solar cells (DSSCs)	Single-junction halide-perovskite (PSCs)	Monolithic perovskite/c-Si tandem (2-terminal)	Commercial crystalline-silicon (c-Si)	Organic solar cells (OSCs)
PCE under 1-sun AM1.5G	15.2% certified PCE for a small-area cell, 10.7% in minimodule and 8.8% in submodule configuration. A semitransparent 400 cm ² module reached 5.1% PCE with 35.7% AVT, illustrating the scale-up penalty but also the multifunctional output [318, 368].	28.0% certified PCE on 0.05 cm ² . The gap between small cells and large-area modules remains relevant, reaching a maximum PCE of 23.9% in minimodule and 22.9% in submodule configuration [379].	35.2% certified PCE on 1 cm ² , and 32.2% for a large (274 cm ²) cell [379].	Commercial modules are typically about 22%–25%, while the certified PCE large-module record is 27.03% [379, 380].	21.01% certified PCE for a single-junction cell. Large flexible modules have reached 14.04% on 41 cm ² and 13.10% on 370 cm ² [381, 382].
Indoor and diffuse light operation	Excellent application-specific response. Devices with 2.8 cm ² active area reached 28.4%–30.2% under ambient light. Dye spectral matching and the mesoporous architecture support favorable diffuse and oblique-light harvesting [318, 383].	Excellent indoor performance. A wide-bandgap PSC reached 37.6% at 1000 Lx. Bandgap tunability is advantageous, although lamp spectrum, calibration, and phase stability remain important [384].	Primarily optimized for AM1.5G. In series-connected two-terminal tandems, spectrum and temperature modify current matching and the optimum top-cell bandgap [385].	Reliable at reduced irradiance, but the fixed Si bandgap limits spectral matching to visible-only artificial sources. Silicon heterojunction devices nevertheless show good low-light behavior [386].	Excellent indoor response. Tunable molecular absorption can be matched to LED spectra; 30.2% at 1000 Lx and 33% under concentrated indoor light have been reported [387].
Operational stability	Strongly architecture-dependent. The 15.2% record cell was tracked for 500 h; semitransparent modules have exceeded 1000 h under ISOS protocols, and an earlier module operated outdoors for approximately 2.5 years. Electrolyte aging, leakage, sealing, corrosion, and dye/electrolyte photochemistry remain key bottlenecks [318, 368, 383].	Rapid progress: a 27.22 cm ² module retained 94.66% after 1000 h of continuous 1-sun illumination. Multiyear outdoor validation and bankable module data remain limited [388].	Recent 1 cm ² tandems achieved T95 >1000 h; a flexible 33.6% tandem showed T80 >2000h under continuous illumination and retained 90% after 1000 h damp-heat [386, 389].	Clear benchmark. Typical service-life assumptions are around 25–30 years, and literature meta-analysis gives median crystalline-Si degradation rates of 0.5–0.6% per year [390].	High-efficiency nonfullerene systems have reached MPP T80 around 1000 h; advanced device engineering has yielded extrapolated T80 >45,000 h, and 7-month outdoor operation has been demonstrated. These results are promising but are not equivalent to multidecade module bankability [391–393].
Mechanical flexibility	Compatible with plastic or metal foils and with low-temperature printed architectures. Flexible	High research-level flexibility: 24.43% PCE with 94.1% retained after 10 000 bending	A certified 33.6% flexible tandem retained 91% after 5000 cycles at a 17.6 mm radius, but the multilayer	Conventional glass-laminated modules are rigid. Edge-blunted, >24%-efficient crystalline-Si cells can	Strongest intrinsic form factor advantage. Ultra-flexible cells reached 18.71%, 30.04 W g ^{−1} , and retained 93.47% after

(Continues)

TABLE 3 | (Continued)

Category	Dye-sensitized solar cells (DSSCs)	Single-junction halide-perovskite (PSCs)	Monolithic perovskite/c-Si tandem (2-terminal)	Commercial crystalline-silicon (c-Si)	Organic solar cells (OSCs)
	modules have been demonstrated, although generally at lower PCE than state-of-the-art flexible PSCs and OSCs [376, 377].	cycles at a 3 mm radius under 65% relative humidity [376].	stack is technologically complex [386].	nevertheless retain their initial PCE after 1000 bending cycles [394].	10,000 cycles at a 1 mm radius; large-area flexible modules are also available [382, 395].
Environmental sustainability and end-of-life	Abundance of TiO ₂ , metal-free dyes, carbon counter-electrodes, aqueous systems, and low-temperature printing can decrease the environmental footprint. However, FTO glass, noble metals (Ag/Pt/Ru), solvents, electrolyte composition, fabrication yield, efficiency, and lifetime remain the main contributors to the overall life-cycle impact. Natural dyes do not necessarily reduce impacts per kWh [396, 397]. Currently, there is no dedicated industrial collection and recycling chain [398].	Thin absorbers and potentially low-temperature manufacturing are promising, but Pb, polar aprotic solvents, In-based TCOs, metal electrodes, process yield, and lifetime remain central uncertainties. Recycling and substrate reuse can markedly improve life-cycle performance [399, 400].	The high energy yield per unit area can compensate for the additional materials required by the tandem architecture. However, the overall environmental footprint reflects the combined impacts of the silicon bottom cell, the perovskite top cell, the TCOs, and the interconnection layers. Furthermore, the lifetime mismatch between the two subcells remains a key challenge [401].	Long lifetime, high efficiency, and mature manufacturing provide low impacts per delivered kWh. Major hotspots include energy-intensive silicon purification/wafer production and Ag use, although material intensity and manufacturing energy continue to decline [39, 402]. High-value silicon recovery remains an active development area [402, 403].	Low material mass, low-temperature printing, and short prospective energy-payback times are attractive. Complex donor/acceptor synthesis, halogenated solvents, ITO/Ag, production yield, and uncertain lifetime can influence strongly life-cycle impact [404]. No established industrial chain yet exists [405].
What makes this technology stand out	A cooptimization of wavelength-selective absorption, color, AVT/CRI, semitransparency, bifaciality, and effective diffuse/indoor harvesting. Particularly attractive for BIPV, greenhouse agrivoltaics, decorative surfaces, and self-powered IoT [318, 368, 383].	Highest single-junction thin-film PCE, strong bandgap and transparency tunability, and excellent indoor potential; long-term stability and Pb management remain decisive [379, 384, 399].	Highest electrical output per unit area; particularly valuable where installation area is constrained and maximum power density is the primary criterion [379].	Robust technology, field-proven lifetime, manufacturing scale, and end-of-life infrastructure [380, 390, 402].	Best combination of ultralight weight, mechanical flexibility, printable area, semitransparency, and molecular spectral tuning. Particularly suited to wearables, lightweight BIPV, portable surfaces, indoor IoT, and visually integrated PV [382, 387, 395, 406].

Notes: "Tandem" refers here to monolithic two-terminal perovskite/c-Si devices. Record-cell, module, and commercial-product efficiencies are not directly equivalent. Indoor PCE depends strongly on lamp spectrum, illuminance-to-irradiance conversion, calibration, aperture definition, and active area. Stability values originate from nonharmonized field and accelerated tests; extrapolated lifetime values are identified explicitly. Life-cycle outcomes depend on the functional unit, fabrication yield, device efficiency, operating lifetime, electricity mix, climate, system boundaries, and recycling assumptions.

outdoor stability, or industrial maturity (mainly due to the liquid nature of the electrolyte and need for sealing). In fact, one of the main barriers to the large-scale commercialization of DSSCs is still their reliance on liquid electrolytes, the volatility of which, risks of leakage and thermal instability complicate long-term sealing and module fabrication. Replacing the liquid phase with quasi-solid or solid-state hole-transporting media is, however, not straightforward. This is because restricted ionic/charge-carrier transport, incomplete penetration into the mesoporous photo-electrode and poor interfacial contact result generally in a trade-off between device stability and photovoltaic performance [73, 144, 408–411]. Their main advantage lies instead in the combination of effective energy harvesting under diffuse, oblique, and low-intensity illumination and a high degree of optical and spectral tunability [412]. Through the molecular design of the sensitizer and the control of the photoelectrode, DSSCs can be tailored in terms of color, transparency, color rendering, and wavelength-selective absorption while maintaining useful photovoltaic performance [413, 414]. This combination is particularly valuable for indoor energy harvesting, transparent BIPV, and greenhouse agrivoltaics, where annual energy yield, visual quality, and control of the transmitted spectrum may be more important than peak efficiency under standard 1-sun conditions. In addition, their compatibility with printable, lightweight, bifacial, and potentially flexible architectures further supports their role in applications that cannot be addressed efficaciously by conventional rigid and opaque photovoltaic modules.

The analysis of the costs of DSSCs in relation to other photovoltaic technologies is generally complicated [415]. In primis it has to be considered the continuous fluctuation of the cost of the energy that is necessary to produce the photovoltaic technologies. In fact, the cost of the energy represents a factor that varies not only on temporal basis due to market fluctuations but also depends also on the origin of the energy, that is, if produced by renewable sources or by fossil fuels, and on the zone of production in dependence of the corresponding local taxation. In addition to the cost of the production energy, it has to be considered the cost of the raw materials and of their synthesis/preparation in their final configuration (sensitizers, substrates, electrodes in the nanostructured version [416] and CE [417]), cost the electrical connection (which can vary even for the same DSSC technology being adoptable either the Z- or W- scheme of configuration for the interconnection of adjacent cells [349, 418–420]), cost of assembly/wiring, and the labor cost. An indicative range of costs for the materials necessary to attain 1 square meter of DSSC is about 20–25 USD considering that about half of the cost is due to the use of TCO-covered glass as substrate [421]. The employment of TCO-covered plastics as substrate [422, 423] could abate the cost of about 30% with respect to glass [424, 425], implying the modification of the scaling-up plant and the procedures of multilayered deposition similar to what has been calculated for perovskite solar cells [426, 427]. Overall, the incidence of the material cost is about 75% on the total cost of a DSSC considering that labor cost is around 10% of the total [421]. Costs of processing stand at about 10% of the total cell cost and are mostly related to the control of temperature and to the definition of the ambient for the cleaning of surfaces. In the process of DSSCs fabrication, the operative step of electrode surface cleaning under controlled conditions of illumination (with UV lamps), chemical composition, and pressure of the cleaning atmosphere (e.g., plasma or ozone), and

ultrasonication prior to the formation of junctions/interfaces is essential to remove chemical contaminants and electronic defects that can cause interfacial resistance between conductors, semiconductors, and photoactive materials [428]. The resistive effect of contaminants (to be considered as point defects) is mainly associated with the creation of surface states that have characteristics of charge trapping/recombination and can consequently block charge transport against the build-up of concentration and electric field gradients that in turn would favor charge transport either for diffusion or for migration during DSSC operation. The temperature of DSSC processing in the phase of material preparation dictates nanoparticle connectivity, material crystallization, and dye surface concentration for the nanostructured semiconducting material, while in the phase of cell assembly, the temperature is crucial for the deposition onto plastic deformable substrates and determines the quality of the adhesion between contacting layers. Control of both temperature and cleaning ambient during fabrication of DSSCs appears then crucial for the optimization and stability of cell conversion performance through the improvement of the charge transport characteristics in the single phase and the properties of charge transfer across interfaces [409]. Consideration of the use of natural dyes as sensitizers could represent a practicable perspective for DSSCs development not only in terms of environmental impact but also in terms of costs with the switch of colorists competences from synthetic chemistry (involving the use of expensive metal-containing precursors and a relatively large consumption of polluting industrial reactants) to extraction/separation chemistry from natural sources with use of not polluting solvents and requiring a generally low level of chemical processability [429]. Moreover, in an indirect way, the adoption of self-powered DSSCs in an indoor ambit governed by IoT for smart climate control and lighting automation can abate the cost of home energy consumption by further 20%–30% with respect to a nonlight-powered system [430, 431].

9 | Conclusions and Perspectives

The current status of the performances achieved with DSSCs, the market potentialities of DSSCs, and the challenges that await a more extended use of DSSCs have been reviewed. In the first instance it has emerged the issue of long-term durability of DSSCs has emerged, an issue that has been tackled through the development of new redox couples surpassing the limitations of the benchmark I_3^-/I^- (these consist of low redox potential, corrosive behavior toward cell components, and intrinsic visible light absorption). Addressing the limitations of conventional redox mediators and developing new systems like those based on Co(III)/Co(II) complexes or Cu(II)/Cu(I) compounds are thus key challenges in bringing DSSCs closer to commercial viability. An advanced class of redox couples is constituted by tetradentate copper cages that have a high rate of interfacial electron exchange and enable values of open-circuit voltage larger than 1.1 V at a single junction. In addition to the development of new redox couples, the increase of DSSC durability has also been tackled with the design of metal-free, fully organic dye-sensitizers, the chemical stability of which is not affected by the weakness of the ligand–metal bond and by the complexity of the electronic transition energy scheme that involves metal-to-ligand charge-transfer processes. At the present level of DSSC electrolyte, the choices are mostly

based on polar liquid systems that suffer from leakage and evaporation and require efficacious sealing systems. In this regard, researchers have considered the use of gel, semisolid, and polymeric electrolytes as well as ionic liquids, deep eutectic solvents, and biopolymers with the challenge of reaching high ionic mobility, high diffusivity for the redox couple, and fast traversing of the electronic charge through the interfaces these semisolid electrolytes of new conception create with the electrodes. Besides the issue of durability, the recent research on dye-sensitizers for DSSCs has also been directed toward the development of sensitizers that present transparency (or quasi-transparency) in the visible spectrum, considering the finality of installing DSSCs for BIPV, agrivoltaics, archaeo-photovoltaics, and smart fenestration while maintaining an acceptable PCE. The guidelines in the design of transparent sensitizers are the achievement of dyes with a correct disposition of the molecular orbitals in order to absorb light in the NIR ($\lambda_{\text{max}} > 750 \text{ nm}$) and/or in the near-UV ($\lambda_{\text{max}} < 380 \text{ nm}$) range. The versatility of organic synthetic chemistry allows for the exploitation of a wide variety of heteroaromatic systems to develop symmetric and asymmetric sensitizers able to target specific spectroscopic features. The insertion of DSSCs in the context of transparent PV introduces new paradigms of quality for DSSCs, like high values of CRI, AVT, and LUE. For this latter parameter, a rule of thumb has been proposed with LUE not smaller than 450 for a practical application of DSSCs with good quality of see-through vision. If indoor PV is the destination of DSSCs, then it must be considered the performance of luminous energy conversion when the incident light passes from the sunlight to an artificial light source. The artificial light sources are characterized by having strong components mostly in the blue, red, or yellow regions with specific peaks in their spectra while lacking considerable emission in the IR and UV ranges. This imposes different criteria of spectral selectivity in the design of dyes of DSSCs for indoor PV with respect to DSSCs for transparent PV. The versatility of synthetic organic chemistry, again, constitutes a useful basis for the preparation of molecules with selected absorption for DSSCs to be employed in indoor PV. Given the possibility of imparting selective absorption to the dyes, another important practical application of DSSCs is the generation of electrical power in a greenhouse and the concomitant manipulation of the light spectrum entering the greenhouse for the optimization of the general and luminous conditions of plant growth and the powering of sensors and irrigation systems. Recently developed classes of DSSC dyes for transparent PV are squaraines, xanthene-based, and aza-BODIPY-containing molecules that are all united by exhibiting NIR absorption and a structural scheme that incorporates donor and acceptor moieties bound together by an extended skeleton with delocalized π -electrons. For advanced transparent DSSCs, it is expected that an advancement of performance through the use of halogen-bonded auxochromes and mixed siloxy/hydroxamate anchors to overcome eventual problems of dye immobilization and poor efficiency of charge transfer. Such problems are due to weak binding of the dye-sensitizer with the semiconducting substrate and nonoptimized electronic coupling between the frontier orbitals of the sensitizer and the band edge levels of the semiconductor. In this regard, a series of dye bases on porphyrins and their thieno-pyrazine derivatives have also been designed and successfully applied with consequent positive impact on long-term device durability. This latter aspect has also been tackled through the approach of interface engineering with the adoption of strategies like the use of blocking layers in contact with the nanostructured

semiconducting substrate or embedding electronically insulating nanoparticles onto the semiconductor substrate to render the process of charge transfer less prone to recombination and energy losses through the shift of the relevant energy levels. The durability tests carried out on DSSCs with passivating layers indicate the possibility of reaching 10 years of operation for indoor PV systems, taking advantage of the high quantum yields of the DSSCs under these conditions. Also, surface functionalization with linkers or systems having high conformational rigidity has been considered for improving charge injection efficiency. CEs, alternative to Pt nanoparticles embedded onto transparent conductive substrates, have also been considered to warrant electrocatalytic properties while being cost-effective: carbon-based materials like nanotubes, graphene, and activated carbon have been studied for the electrocatalysis of reduction reactions at the cathodic CE. Also, inorganic layered materials have been characterized as CE for DSSCs. Among these, the most promising examples are the transition-metal dichalcogenides MoS_2 and WS_2 . Other electrodic materials for DSSCs of the organic polymeric type are PEDOT:PSS and polyaniline because of good catalytic activity toward a variety of redox mediators and, more importantly, because they offer the possibility of being processed also on flexible substrates at relatively low temperatures of deposition while offering good adhesion.

In terms of scalability and industrial manufacturing of DSSCs, one of the most critical questions is the attainment of large-scale uniformity in the phase of preparation. Another critical issue is the reproducibility of the conversion performances for the DSSCs in the assembled final version. Given the multilayered nature of a DSSC, it has been early recognized that screen printing represents an ideal technique for industrial manufacturing and that continuous optimization of screen printing conditions (including tight control of printing parameters and precise definition of ink formulation) is necessary to overcome questions of uniformity of the coverage in the various layers of the device as well as problems of performance reproducibility. Besides uniformity of the various layers constituting the single cell, another concern is the uniformity of the deposition in a solar DSSM, (a system containing several cells on the same supporting layer and representing the actual unit of the scaling-up process). The increase in module sizes requires a careful engineering process to prevent performance loss while realizing complex schemes of electrical connectivity. At the industrial level, the use of liquid electrolytes in DSSCs requires adequate sealing to prevent electrolyte evaporation and moisture ingress, which both contribute to the acceleration of the degradation. For that, the development of suitable sealants and the use of materials that withstand temperatures of deposition/assembly are crucial for the industrialization of DSSCs production and render viable the commercialization of DSSCs. In the passage from laboratory to industrial scale, the most demanding challenges are optimization of materials and enlargement of the photoactive area while maintaining the overall conversion efficiency at levels not lower than 10% when the area is at least 50 cm^2 . In this regard, the automation with pilot fabrication lines for the manufacturing processes will warrant repeatability in the photoconversion performance and precision in the DSSCs construction to advance this PV technology toward its commercialization in the most disparate directions of application.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

Data that have been produced in our laboratories will be made available upon request.

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