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Review

Comparison of Hybrid and Single Dyes for DSSCs: Fabrication, Efficiency, and Economic Evaluation

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Abstract: As the global community needs sustainable alternatives to silicon-based photovoltaics, dye-sensitized solar cells (DSSCs) have been regarded as a third-generation technology due to their low-cost fabrication and tunable aesthetics. However, the efficiency of single-sensitizer systems, including high-cost ruthenium complexes and low-stability natural pigments, remains constrained by narrow absorption spectra and rapid charge recombination. The transition from single to hybrid dye systems highlights the technical and economic necessity of multi-chromophore and metal-doped architectures. By integrating previous results on silver doping and cocktail sensitization, hybrid dye systems achieve panchromatic absorption and a significant reduction in the semiconductor bandgap by up to 3.0 eV. The results of this review provide a reference for the development of next-generation sensitizers that offer industrial-grade stability and a lower levelized cost of electricity, facilitating the integration of DSSCs into building-integrated photovoltaics and portable electronics.

Keywords: Dye-sensitized solar cells (DSSCs), Co-sensitization, Panchromatic absorption, Electrochemical impedance spectroscopy (EIS), Power conversion efficiency (PCE), Economic evaluation

1. Introduction

The accelerating global energy demand and the detrimental environmental effects of fossil fuels have intensified the requirements for renewable energy. While conventional silicon-based solar cells are widely used, their implementation is constrained by high manufacturing costs, energy-intensive production processes, and mechanical rigidity. These also limit the integration of silicon-based solar cells into flexible and lightweight applications [1]. To address the limitations, dye-sensitized solar cells (DSSCs) have been developed and implemented as a third-generation photovoltaic technology, offering advantages such as facile fabrication, transparency, tunable coloration, and efficient operation even under low-light and indoor conditions. Such characteristics enable DSSCs to be available for building-integrated photovoltaics (BIPV) and portable electronic devices [2].

A typical DSSC consists of a transparent conductive oxide (TCO) substrate, a nanocrystalline semiconductor (commonly titanium oxide (TiO_2)), a photosensitizer (dye), a redox electrolyte, and a counter electrode. The sensitizer plays an essential role in energy-harvesting as it absorbs photons and injects electrons into the TiO_2 conduction band, initiating the photo-electrochemical process that mimics natural photosynthesis [3]. Since their seminal introduction by O'Regan and Grätzel [4], DSSCs have attracted considerable attention as cost-effective and environmentally benign alternatives to silicon photovoltaics.

In the fabrication of DSSCs, the selection of dye is critical as the dye determines the absorption spectrum, electron injection efficiency, and overall device performance. Recently, ruthenium-based complexes such as N3 and N719 have been used as they achieve power conversion efficiencies exceeding 11–12% [5]. However, their widespread adoption is hindered by the high cost of noble metals, complex synthesis routes, and concerns regarding toxicity and environmental pollution. Consequently, natural dyes derived from plants have been researched as sustainable, non-toxic, and inexpensive alternatives, as using natural dyes satisfies the requirements of green chemistry and circular economy [6,7]. However, despite their ecological advantages, natural dyes have limited stability and narrower absorption ranges, necessitating extensive optimization.

To overcome these limitations, dye mixtures (hybrid dyes) and their co-sensitization strategies have been proposed. In hybrid dyes, organic push–pull (D–π–A) dyes are combined with metal complexes or employ multi-chromophore assemblies to enhance panchromatic absorption and enhanced stability [8,9]. Such combinations improve the spectral response, charge transfer dynamics, and reduce recombination losses, increasing DSSC efficiency and durability.

Given the rapid progress in dye selection and design, and the increasing emphasis on sustainable energy use, a comprehensive review of single and hybrid dye systems is required. Therefore, a review is conducted to evaluate the structural, photophysical, and electrochemical properties of the hybrid and single dyes, highlighting their roles in developing and manufacturing DSSCs. By examining the interplay between molecular anchoring, fabrication complexity, and economic feasibility, the results of the review contribute to the rational design of next-generation sensitizers and the wider adoption of DSSCs in renewable energy production.

2. Technical Perspectives

2.1 Electrochemical Impedance Spectroscopy (EIS) and Kinetics

Electrochemical impedance spectroscopy (EIS) is a primary tool for elucidating the performance differences between single and hybrid dye systems. The scientific and technical parameters of the hybrid and single dye systems are compared in Table 1.

Table 1. Comparison of scientific and technical parameters of hybrid and single dye systems.

Feature	Single dye (anthocyanin)	Hybrid dye (ABC/Ag)	Advantage
PCE (%)	0.12–0.26%	0.60–0.74%	Synergistic photon harvesting [6]
Bandgap (Eg)	–3.2 eV	Reduced by –3.0 eV	Easier electron excitation [6]
Stability	Low (hydration/pH sensitive)	High (via co-pigmentation)	Protected chromophore nucleus [7]
Recombination	High (surface voids)	Low (denser monolayer)	Suppressed back-electron transfer [8]
Conductivity	Moderate	0.534 $\Omega^{-1}m^{-1}$	Enhanced charge transport [9]

In synthetic benchmarks and natural dye mixtures, performance enhancement is frequently attributed to reduced recombination and improved charge transport [3]. In single-sensitizer systems, the dye layer contains interstitial voids that expose the TiO_2 surface to the electrolyte, facilitating back-electron transfer. Hybrid dyes, such as synthetic co-sensitizers (D205) or natural dye mixtures, create a denser molecular monolayer than single dyes that physically shields the TiO_2 surface and suppresses recombination [8]. This structural modification increases the recombination resistance and directly improves device stability.

The synergistic effect of hybrid systems also extends the electron lifetime by reducing the density of surface trap states. For example, hybridizing anthocyanin with chlorophyll or betalain widens the absorption spectrum and improves conversion efficiency, enhancing performance from 0.125% for single dyes to 0.602% for hybrid mixtures [6]. Incorporating metal ions such as silver (Ag) into hybrid dye mixtures leads to optimizing kinetics by providing conductive pathways and inhibiting rutile phase formation in TiO_2, thereby facilitating faster charge transfer [9]. One of the most significant advantages of hybrid dyes, particularly when doped with metals, is their ability to reduce the bandgap of the TiO_2 photoanode. The hybrids of anthocyanin, betalain, and chlorophyll (ABC) doped with Ag reduce the TiO_2 bandgap by 3.0 eV, encouraging electron excitation and improving charge injection [6]. Hybrid dye solutions exhibit higher conductivity than single dyes. For instance, the ABC/Ag hybrid system presented a peak conductivity of 0.534 $\Omega-1m-1$, significantly enhancing the transport of photo-generated electrons [9].

A hybrid dye system requires complex molecular mechanisms that extend beyond simple light absorption. In single natural dyes such as anthocyanin, molecules are highly susceptible to hydration, which leads to the formation of colorless carbinol pseudo-bases and rapid degradation. Hybridization through co-pigmentation stabilizes the chromophore by blocking hydration through vertical π–π stacking between planar dye structures [6]. This mechanism maintains the flavylium cation form of anthocyanin, particularly under acidic conditions, and ensures consistent electron injection [7]. Hybridization also prevents aggregation, which is a common issue in flat-structured dyes such as porphyrins and tends to self-quench when stacked on TiO_2 surfaces. The introduction of auxiliary chromophores or spacer molecules mitigates this aggregation and maintains efficient electron injection [9].

Another advantage of hybrid systems is the cocktail effect. The combination of dyes with complementary absorption spectra enables synergistic photon harvesting. Anthocyanin absorbs light in the 300–400 and 500–600 nm wavelength, while chlorophyll absorbs light in the 600–700 nm wavelength. Their combination ensures photon harvesting across the entire visible spectrum (400–800 nm), which is rarely absorbed by using single natural or organic dyes [6]. This spectral broadening, coupled with enhanced stability through acidified solvents such as citric acid, presents the superior performance of hybrid dye systems [7].

2.2 Transition from Single to Hybrid Dye Systems

The transition from single to hybrid dye systems addresses two problems in DSSC technology: the narrow spectral response and poor long-term stability. The synergistic effect of hybridization is presented by conducting incident photon-to-current efficiency (IPCE) measurements and thermal aging tests and highlighting the advantages of combining multiple sensitizers.

Single-dye systems, such as N719 or pure anthocyanin, exhibit IPCE spectra limited by the specific absorption bands of the chromophore. For example, N719 peaks at approximately 530 nm but declines sharply beyond 600 nm, while natural dyes such as chlorophyll capture only the red and blue regions, leaving a pronounced green gap in the absorption profile [10]. Hybrid systems, such as ABC mixtures, show a more panchromatic response by broadening the absorption spectrum significantly more than individual components [10]. When these hybrid mixtures are doped with Ag nanoparticles, conductivity significantly increases (up to 0.534 Ω−1m−1), facilitating efficient charge collection across the broadened spectral range [10].

A barrier to DSSC commercialization is the desorption of dyes under thermal and environmental stress. The stability of the dye–semiconductor interface is governed by the chemical nature of the anchoring groups. Synthetic single dyes that rely solely on carboxylic acid (−COOH) anchors are prone to hydrolysis of ester bonds under thermal stress at approximately 60°C. In contrast, hybrid systems incorporating co-sensitizers with phosphonic acid (−PO3H2) anchors demonstrate excellent durability. The formation of P–O–Ti bonds enables a molecular seal to maintain 94% of the initial PCE after 1,000 hours of testing, compared with a 22% drop in single −COOH systems [11]. Natural hybrids' stability is largely affected by the anthocyanin structure. Anthocyanins maintain stable flavylium cation forms only under acidic conditions. Acidified extraction solvents, such as citric acid, are used as chemical stabilizers to prevent the transition to the unstable colorless carbinol pseudo-base [10]. Co-pigmentation occurs as a natural form of hybridization when anthocyanins associate with other organic molecules and shield the chromophore from nucleophilic attack by water, enhancing photostability [10] (Table 2).

Table 2. Comparison of stability and kinetic parameters of hybrid and single dye systems.

Dye system	Dominant anchor	Recombination resistance (Ω)	Lifetime (ms)	PCE retention time (1,000 hrs)
Single synthetic (N719)	−COOH	98	12.4	Up to 78%
Hybrid synthetic	−COOH/−PO3H2	154	18.7	Up to 94%
Single natural (A)	−OH	Low	Up to 5.2	-
Hybrid natural (ABC/Ag)	−OH / metallic	High	Up to 14.5	Up to 85% (with stabilizers)

EIS analysis results reveal that hybrid systems alter the kinetics at the TiO_2/dye/electrolyte interface. The hybrid dye system extends electrons' lifetimes from 12.4 to 18.7 ms in co-sensitized cells. Such an extension of the lifetime increases the open-circuit voltage, as secondary dye molecules fill surface trap states and prevent recombination with triiodide (I3−) ions in the electrolyte [7].

2.3 Light Conversion Efficiency

The metric for evaluating DSSCs is power conversion efficiency (PCE), which depends on the dye's ability to harvest photons and inject electrons into the semiconductor. Hybridization outperforms single-dye systems by addressing spectral limitations and charge transfer bottlenecks through synthetic co-sensitization or natural pigment blending [10]. In synthetic sensitizers, ruthenium complexes such as N3 and N719 are regarded as the benchmark and show 11–12% light conversion efficiencies owing to their stable metal-to-ligand charge-transfer transitions [5]. Organic single dyes show lower efficiencies of 8–10% [12]. Hybrid dye systems surpass such efficiency by utilizing multiple chromophores to create a cocktail effect, which broadens the absorption spectrum and reduces recombination at the TiO_2 interface. These hybrid architectures increase the efficiency to 13–14%, which is higher than that of single-dye systems [8].

The performance improvement in hybrid dye systems is achieved by several factors. First, synergistic spectral coverage plays an important role. Single dyes often exhibit narrow absorption bands; for example, chlorophyll absorbs primarily in the red and blue regions but lacks absorption in the green region. ABC creates a panchromatic response, increasing conversion efficiency from 0.125% for single chlorophyll to 0.602% for the hybrid ABC mixture [6]. Second, the metal doping in the hybrid dye system enhances conductivity and bandgap engineering. The integration of Ag nanoparticles into hybrid dye mixtures (e.g., ABC/Ag) increases the conductivity of the dye layer to 0.534 Ω−1m−1 and reduces the TiO_2 photoanode bandgap. These modifications resulted in a peak efficiency of 0.742% [7]. Finally, acidification enables the equilibrium of natural dyes by making stable flavylium cation forms. In anthocyanin-based cells, acidified solvents such as hydrochloric acid facilitate efficient electron transfer to the TiO_2 conduction band, which increases PCE from 0.64 to 0.84% [7]. This chemical stabilization underscores the importance of solvent engineering in natural hybrid systems.

3. Fabrication Methods

In the fabrication of high-performance dye-sensitized solar cells (DSSCs), the quality of the molecular monolayer and the efficiency of charge transfer are critical determinants of device performance. The fabrication process varies considerably between single natural dyes, metal-doped hybrid systems, and synthetic benchmarks. For natural pigments, performance is heavily dependent on the extraction process and the chemical environment created by solvents.

Anthocyanin is commonly extracted from sources such as dragon fruit peel using a mixture of methanol, acetic acid, and distilled water in a volumetric ratio of 25:4:21. Acidification is essential in this process, as it stabilizes the flavylium cation, the red-colored form most effective for light harvesting [11]. Betalain is mainly sourced from Bougainvillea glabra flowers by immersing the petals in a distilled water–acetone mixture (2:1 v/v) for 24 hours [13]. Chlorophyll is generally extracted from cassava leaves using an acetone–ethanol mixture in a 1:5 ratio [14]. Solvent selection plays an essential role in these protocols. Polar solvents such as ethanol and methanol are preferred for anthocyanins, but their concentration and pH must be carefully controlled to prevent pigment degradation into colorless carbinol pseudo-bases or chalcones. Hybrid dye systems are fabricated by mixing individual dye solutions in specific ratios, often 1:1 or 1:1:1, to achieve a cocktail effect that broadens spectral coverage. Recent innovations highlight the doping of hybrid mixtures with silver nanoparticles. Ag doping improves conductivity and reduces the TiO_2 photoanode bandgap. In addition, silver inhibits the formation of the rutile phase in TiO_2, fostering efficient charge transfer and suppressing recombination [10]. Synthetic single dyes, such as ruthenium complexes, are produced via conventional coordination chemistry. In contrast, hybrid architectures require multi-step synthesis, which involves covalent linking of chromophores or the incorporation of metal–organic frameworks. This added complexity reflects the trade-off between fabrication difficulty and performance enhancement.

The method of adsorbing the dye onto the TiO_2 surface is different for the DSSC fabrication in the hybrid or single dye systems. In the hybrid system, the TiO_2 film is immersed in a mixed dye solution, and the molar ratios must be carefully adjusted to 10:1 or 20:1, to account for the differing binding kinetics of the pigments. In sequential adsorption, the photoanode is sensitized with a dye of high binding affinity, such as a metal complex, for 12 hours, which is followed by immersion in a second dye, typically an organic pigment, for 2–6 hours. Sequential adsorption is used to avoid competitive displacement and ensures the formation of a denser, uniform monolayer, enhancing both stability and efficiency [15].

4. Economic Evaluation

The cost-effectiveness of DSSCs is analyzed by using a model that accounts for raw materials, substrates, electrolytes, and labor for manufacturing. For single-dye modules, the raw material cost is approximately USD 12.50/m2, while hybrid dye systems when prepared from mixed natural pigments, reduce the cost to USD 9.80/m2 owing to the lower cost of blended organic dyes [11]. The cost of TiO_2 and fluorine-doped tin oxide glass remains constant at USD 45.00/m2 in both systems, while electrolytes and sealants contribute an additional USD 15.00. Labor costs differ significantly: single-dye systems require simpler dipping processes (USD 8.00/m2), whereas hybrid systems involve sequential adsorption or multi-step fabrication, raising labor costs to USD 14.00/m2 [15]. Consequently, the total capital expenditure is slightly higher for hybrid dye systems (USD 83.80/m^2) than for single-dye systems (USD 80.50/m^2).

Despite the USD 3.30/m^2 increase in fabrication costs for hybrid dye systems, the 27% increase in power output lowers the levelized cost of electricity (LCOE), which is defined as (total life cycle cost)/(total energy generated). For single-dye DSSCs, the LCOE is approximately USD 0.085/kWh, whereas hybrid systems require an average LCOE of USD 0.069/kWh [6]. This reduction makes hybrid dye systems for DSSCs attractive for BIPV, where the cost of the substrate (glass) is fixed, and maximizing efficiency per square meter is the primary driver for return on investment.

Single dyes, particularly ruthenium complexes, are expensive due to their scarcity and the complexity of synthesis [5]. Organic single dyes are cheaper but suffer from lower stability and reduced long-term viability [12]. Hybrid dyes, by contrast, involve higher synthesis costs due to fabrication complexity, especially when incorporating metal nanoparticles or multi-chromophore systems [10]. However, these costs are complemented by long-term economic benefits if stability and efficiency gains are ensured. For instance, Ag-doped hybrid systems reduce the TiO_2 bandgap and improve conductivity, enhancing device durability and lowering operational costs over time [16].

5. Conclusions

Sensitizer materials have been diversified from synthetic ruthenium complexes to organic push-pull dyes and natural plant extracts. This review is conducted to examine previous research results to evaluate DSSC performance by comparing peak efficiency, long-term stability, and economic feasibility of the hybrid and single dye systems. As energy demands increase, it is essential to understand the relationship between molecular anchoring and interfacial kinetics to apply DSSC technology to industrial-scale applications. Synergies from Ag-doping and cocktail effect are important drivers for the next generation of renewable energy

harvesting. The integration of Ag nanoparticles into hybrid natural dyes increases conductivity, which enhances charge transport in solar cells. Specific fabrication methods, such as sequential adsorption, mitigate competitive displacement, ensure a denser molecular monolayer that is essential for high-performance energy harvesting. While single-dye systems have long been researched, hybrid systems show their technical and scientific superiority. Single dyes such as N719 or chlorophyll exhibit absorption gaps (the green gap for chlorophyll), while hybrid ABC mixtures provide a panchromatic response, capturing photons across the entire 400–800 nm visible spectrum. Hybrid systems utilize co-sensitizers with robust anchoring groups, such as phosphonic acid, which form stronger P–O–Ti bonds compared with -COOH anchors used in many single dyes. This results in a 94% PCE retention after 1,000 hours, vastly superior to the 22% reduction in single-anchor systems.

Although fabrication costs for hybrid modules are slightly higher (USD 83.80/m² compared with USD 80.50/m² of single-dye systems), the 27% increase in power output lowers the LCOE to USD 0.069/kWh. This makes hybrid systems the superior choice for maximizing return on investment in BIPV applications.

The evolution of DSSC technology is related to the design of efficient and stable photosensitizers. Hybrid dye systems represent a transition to a new method through synergistic photon harvesting and metal-doping. They also resolve the limitations of narrow spectral response and poor thermal stability that have historically hindered natural and organic DSSCs. Hybrid natural dyes, particularly those doped with Ag, facilitate easier electron excitation by reducing the semiconductor bandgap while simultaneously providing a molecular seal against recombination. Despite the added synthesis complexity, the lower cost of electricity and high PCE retention of hybrid systems confirm their readiness for large-scale renewable energy applications. However, it is necessary to optimize the ratios of cocktail mixtures to refine the balance between cost and panchromatic efficiency.

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