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Review Article

Recent advances in co-sensitized natural and synthetic dye-sensitized solar cells: A critical review

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ABSTRACT

Dye-sensitized solar cells (DSSCs) have garnered significant attention as an environmentally friendly and cost-effective alternative to conventional photovoltaic technologies. This critical review focuses on recent developments in co-sensitized DSSCs incorporating both natural and synthetic dyes. We discuss material selection, co-sensitization mechanisms, device fabrication methods, performance enhancement strategies, stability improvements, and future research directions. The review highlights how the combination of natural dyes, such as Hibiscus and Anthocyanin extracts, with synthetic dyes, including ruthenium complexes and metal-free organic dyes, improves light absorption, charge transfer, and overall device efficiency. Strategies to optimize TiO_2 photoanode morphology, electrolyte composition, and dye loading are elaborated. Finally, potential industrial applications and environmental benefits of co-sensitized DSSCs are discussed.

1. Introduction

Dye-sensitized solar cells (DSSCs) were first introduced by O'Regan and Grätzel in 1991 [1], marking a paradigm shift in third-generation photovoltaic technologies. Unlike conventional silicon-based solar cells, DSSCs offer advantages such as low-cost materials, facile fabrication, semi-transparency, and environmental compatibility. DSSCs generally consist of a dye-sensitized nanostructured TiO_2 photoanode, a redox electrolyte, and a counter electrode. Light absorption by dye molecules leads to electron injection into the TiO_2 conduction band, which then travels to the external circuit while the dye is regenerated by the electrolyte [2, 3].

Recent trends have focused on improving DSSC efficiency and stability by employing co-sensitization strategies that combine two or more dyes with complementary absorption spectra. Co-sensitization can significantly broaden the spectral response, enhance photon harvesting, and reduce electron-hole recombination. This approach is particularly effective when combining natural and synthetic dyes, leveraging sustainability while maintaining high efficiency [4, 5].

Natural dyes derived from plants, fruits, and vegetables, such as Hibiscus, Beetroot, Anthocyanin, and Chlorophyll extracts, offer low-cost, non-toxic alternatives. However, their narrow absorption ranges and lower chemical stability limit device performance. Synthetic dyes, particularly ruthenium-based complexes and metal-free organic dyes, offer high molar extinction coefficients, broad absorption, and superior stability [6–8]. Co-sensitization strategies aim to combine the advantages of both dye types, resulting in higher photocurrent densities (Jsc), improved open-circuit voltages (Voc), and enhanced overall efficiency [9].

This review critically evaluates the literature on co-sensitized DSSCs, focusing on natural-synthetic dye combinations, device optimization, charge transfer mechanisms, stability enhancement, and future research directions.

2. Natural dye-based DSSCs

Natural dyes are predominantly composed of pigments such as anthocyanins, betalains, chlorophylls, and carotenoids. These dyes offer environmental benefits and ease of extraction using solvents like ethanol, methanol, acetone, or aqueous solutions. UV-Vis spectroscopy is commonly employed to analyze absorption maxima, which typically range from 400–600 nm, depending on the dye type [10, 11].

A. Extraction and Characterization

Extraction techniques involve maceration, Soxhlet extraction, or ultrasonication to obtain concentrated pigment solutions. Stabilization of natural dyes can be achieved through pH adjustment, metal ion complexation (e.g., Al^{3+}, Mg^{2+}), and encapsulation in biopolymers. Characterization includes absorption spectra, fluorescence properties, and redox potential determination to assess suitability for DSSC applications [12].

B. Performance in DSSCs

Natural dye-based DSSCs typically demonstrate efficiencies ranging from 0.5% to 4.5%. Performance can be improved via co-sensitization or chemical modification to extend absorption and increase electron injection efficiency. For example, co-sensitization of Hibiscus with Bromophenol Blue has shown an increase in Jsc and overall efficiency due to complementary absorption peaks [13, 14].

C. Challenges

Despite environmental advantages, natural dyes face challenges: photodegradation under prolonged illumination, lower electron injection efficiency, and instability in aqueous electrolytes. Addressing these challenges requires optimized device engineering and co-sensitization strategies.

3. Synthetic dye-based DSSCs

Synthetic dyes, including ruthenium polypyridyl complexes and metal-free organic dyes, are employed to achieve high efficiency in DSSCs. Ruthenium-based dyes exhibit broad absorption (400–700 nm), high molar extinction coefficients, and long-term stability, making them the benchmark for high-performance devices [15, 16].

A. Organic Metal-Free Dyes

Donor–π–acceptor (D–π–A) structured dyes provide an alternative to metal complexes. Their energy levels can be engineered for efficient electron injection and strong absorption in the visible region. Metal-free dyes offer advantages such as low cost, non-toxicity, and structural tunability [17, 18].

B. Limitations

High synthesis cost, purification requirements, and potential toxicity are limitations for synthetic dyes. Furthermore, narrow absorption windows in some organic dyes limit full-spectrum photon harvesting, necessitating co-sensitization to enhance light capture.

4. Co-sensitization strategies

Co-sensitization combines two or more dyes with complementary absorption spectra to enhance light harvesting and reduce recombination. Two primary approaches are employed:

A. Natural-Natural Dye Co-Sensitization

Combining natural dyes, such as Anthocyanin with Chlorophyll extracts, improves absorption across the visible spectrum, leading to higher photocurrent densities. Optimized ratios and sequential adsorption methods prevent competitive adsorption and ensure effective energy transfer [19].

B. Natural-Synthetic Dye Co-Sensitization

Pairing a natural dye with a synthetic dye leverages sustainability while maintaining high efficiency. For instance, combining Hibiscus with a ruthenium-based dye extends absorption range and enhances electron injection efficiency. Sequential and co-adsorption techniques are employed to control dye coverage and prevent aggregation [20].

C. Performance Metrics

Co-sensitized DSSCs demonstrate higher Jsc, Voc, and fill factor (FF) compared to single-dye cells. Electrochemical impedance spectroscopy (EIS) reveals reduced recombination resistance and increased electron lifetime, contributing to higher device stability and efficiency.

5. Device fabrication techniques

Device performance depends on fabrication methods for each component:

TiO_2 Photoanode: Nanoparticle paste deposition via doctor-blade, screen printing, or hydrothermal growth. Morphology and thickness optimization influence electron transport and light absorption.

Counter Electrode: Platinum-coated FTO, carbon-based, or conductive polymer electrodes. Surface area and conductivity impact electron transfer kinetics.

Electrolyte: Liquid iodide/triiodide, cobalt complexes, or solid-state hole transport materials. Composition affects redox regeneration and device stability.

Dye Loading: Immersion time, concentration, and co-sensitization ratio are critical for uniform coverage and optimized light absorption.

6. Electron transfer mechanism

Upon photon absorption, electrons are excited from dye HOMO to LUMO and injected into the TiO_2 conduction band. The electrons then travel to the FTO electrode, while the dye is regenerated via the redox electrolyte. Co-sensitization reduces recombination, optimizes energy alignment, and prolongs electron lifetime, improving photovoltaic performance.

7. Performance optimization

Optimization strategies include:

1. Controlling TiO_2 thickness and nanostructure to balance light harvesting and charge transport.
2. Fine-tuning dye concentrations, adsorption time, and co-sensitization ratios.
3. Electrolyte composition modification to reduce recombination and enhance long-term stability.
4. Incorporating additives or surface modifiers to enhance interfacial charge transfer.
5. These strategies can increase DSSC efficiency by 20–50% compared to single-dye systems.

8. Advanced characterization techniques

Advanced characterization plays a crucial role in understanding the internal physics of co-sensitized DSSCs. Techniques such as Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy (TEM) are used to analyze TiO_2 morphology and porosity. X-ray diffraction (XRD) determines crystallinity and phase composition of TiO_2 (anatase or rutile), which significantly affects electron transport. UV–Visible spectroscopy evaluates dye absorption spectra and confirms successful co-sensitization through broadened spectral response.

Electrochemical Impedance Spectroscopy (EIS) is widely used to analyze charge transfer resistance, recombination kinetics, and electron lifetime. Nyquist and Bode plots provide insight into interfacial charge transport processes. Incident Photon-to-Current Conversion Efficiency (IPCE) measurements further validate wavelength-dependent photocurrent enhancement in co-sensitized systems.

9. Comparative analysis of single vs co-sensitized DSSCs

Single dye DSSCs often suffer from limited spectral absorption and recombination losses. In contrast, co-sensitized DSSCs exhibit:

- Broader absorption spectrum (400–750 nm range)
- Increased short-circuit current density (Jsc)
- Improved power conversion efficiency (PCE)
- Better surface coverage of TiO_2 nanoparticles

However, improper dye ratios may lead to competitive adsorption and reduced performance. Therefore, optimization of dye concentration and sequential adsorption techniques is essential.

10. Economic and commercial considerations

For large-scale commercialization, cost analysis is essential. Natural dyes significantly reduce material costs compared to ruthenium-based dyes. Carbon-based counter electrodes further reduce dependency on expensive platinum. Roll-to-roll fabrication and flexible substrates can enable scalable production. Building-integrated photovoltaics (BIPV), indoor energy harvesting devices, and portable electronics represent promising application areas for co-sensitized DSSCs.

11. Environmental impact assessment

Co-sensitized DSSCs align with sustainable development goals by reducing toxic metal usage and lowering carbon footprint. Life cycle assessment (LCA) studies suggest DSSCs require less energy input during fabrication compared to crystalline silicon solar cells. Biodegradable natural dyes further enhance environmental compatibility.

12. Research gaps and recommendations

Despite progress, several research gaps remain:

1. Long-term outdoor stability studies exceeding 5–10 years.
2. Development of fully solid-state, leakage-free electrolytes.
3. Improved encapsulation techniques for moisture resistance.
4. Integration with hybrid photovoltaic architectures.
5. Standardization of testing protocols for co-sensitized systems.

Future research should emphasize interdisciplinary approaches combining material science, nanotechnology, and device engineering to achieve efficiencies above 15% with long-term durability.

13. Figures and tables

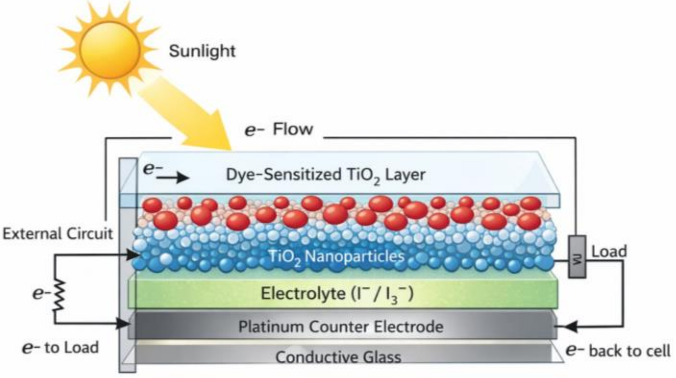

Figure 1: Schematic of DSSC Structure.

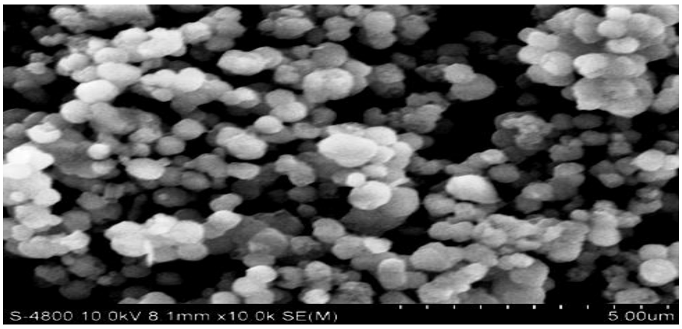

Figure 2: SEM Image of TiO_2 Nanostructure.

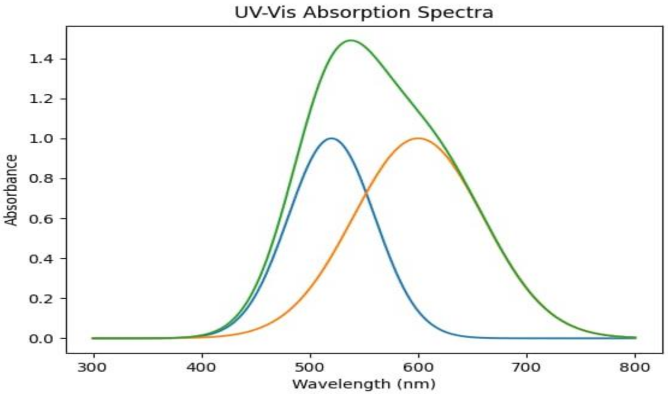

Figure 3: UV–Vis Absorption Spectra.
(Plot comparing absorption of natural dye, synthetic dye, and co-sensitized system showing broadened spectrum.)

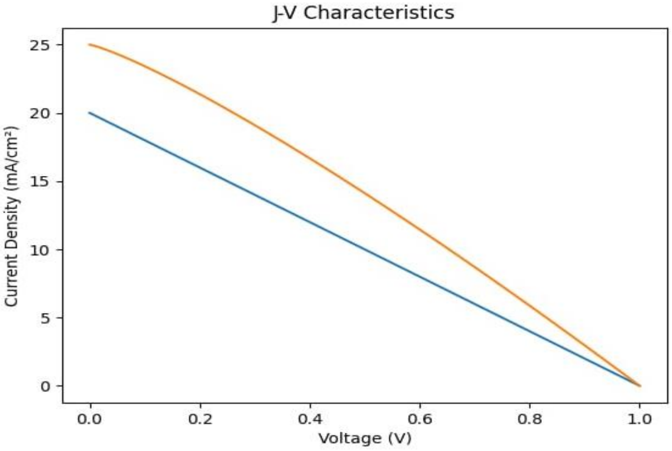

Figure 4: J–V Characteristics under AM 1.5G (100 mW/cm^2).
(Plot of current density vs voltage for single-dye vs co-sensitized DSSCs indicating improved Jsc and PCE.)

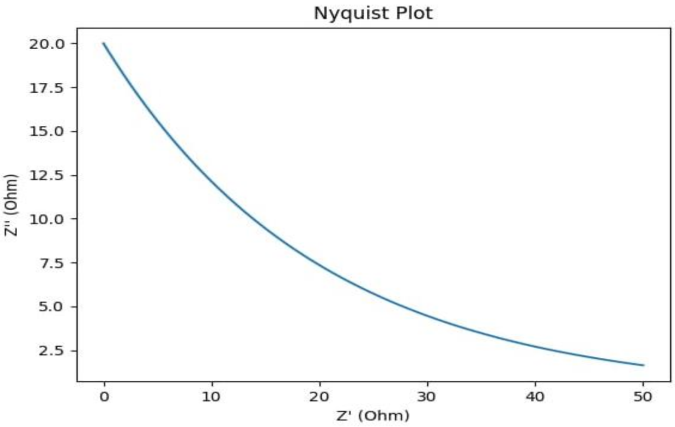

Figure 5: Nyquist Plot from EIS Analysis.

Table 1: Photovoltaic Performance Comparison.

Parameter	Natural Dye DSSC	Synthetic Dye DSSC	Co-Sensitized DSSC
J_{sc} (mA/cm²)	4.5–8.0	12–20	15–22
Voc (V)	0.5–0.65	0.65–0.85	0.7–0.9
Fill Factor (%)	50–60	60–75	65–78
Efficiency (%)	1–4.5	7–14	9–16

Table 2: Effect of Co-Sensitization Parameters.

Parameter	Effect on Performance
Dye Ratio	Optimized ratio increases light absorption and reduces recombination
Adsorption Time	Longer time improves dye loading but may cause aggregation
TiO_2 Thickness	Higher thickness increases absorption but may reduce electron transport
Electrolyte Type	Impacts dye regeneration and stability

Table 3: Comparison of Electrolytes.

Electrolyte Type	Advantages	Limitations
Iodide/Triiodide	Stable, widely used	Volatility, leakage
Cobalt Complex	Higher Voc	Cost, stability issues
Solid-State HTM	No leakage	Lower conductivity

14. Stability and environmental considerations

Long-term stability remains a key challenge for DSSCs, particularly with natural dyes. Techniques such as encapsulation, electrolyte additives, UV filters, and solid-state hole transport materials improve stability. Co-sensitized DSSCs also provide environmental advantages by reducing the amount of synthetic dye required while maintaining high performance, making them suitable for sustainable energy applications.

15. Future prospects

Development of highly stable natural dyes with extended absorption and improved photostability. Integration of co-sensitized DSSCs with tandem or perovskite solar cells for enhanced efficiency. Exploration of solid-state electrolytes and hybrid charge transport layers to increase device lifetime. Scalable fabrication techniques for industrial application, including roll-to-roll printing and flexible substrates. Further studies on device encapsulation, degradation mechanisms, and outdoor performance for commercialization.

16. Conclusions

Co-sensitized DSSCs combining natural and synthetic dyes present a promising approach to achieve high efficiency, low-cost, and environmentally sustainable solar energy devices. Optimized dye selection, device fabrication, and performance enhancement strategies can overcome the limitations of single-dye systems. The combination of eco-friendly natural dyes with high-efficiency synthetic dyes enables broader absorption, improved charge transfer, and enhanced device stability. Future research should focus on long-term durability, scalable fabrication, and integration into hybrid photovoltaic systems.

Authors' contributions

All authors contributed equally to the conception, design, experimental work, data analysis, interpretation of results, and preparation of the manuscript. All authors reviewed and approved the final version of the manuscript for publication.

Conflicts of interest

The author declares no conflict of interest.

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Data availability

No new data were created.

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