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Unlocking the potential of anthocyanin-based dye-sensitized solar cells: Strategies for enhancing efficiency, stability, and economic viability

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Abstract: The primary objective of this research endeavor is to assess the economic feasibility, stability, and efficiency of anthocyanins as natural dyes utilized in dye-sensitized solar cells (DSSCs) as environmentally sustainable substitutes within the realm of renewable energy. Despite anthocyanins offering cost-effective controlled modification of optical properties, challenges persist in achieving high efficiency, long-term stability, and commercial affordability. To address these challenges, a comprehensive research methodology involves anthocyanin extraction from diverse plant sources. The study systematically analyzes the impact of anthocyanins on DSSC performance through precise optical property assessments. Strategic optimization techniques, including dye mixing, co-pigmentation, and modifications to anthocyanin dye structures, are explored to enhance overall solar cell efficiency. Additionally, the investigation includes the application of protective coatings to improve long-term stability. Integral to the research are economic considerations, availability, and scalability, emphasizing the practicality and potential large-scale implementation of anthocyanin-based DSSCs. Innovative approaches, such as dye engineering and novel device architectures, are introduced, reflecting a forward-thinking perspective to address existing challenges. The study uses novel optimization methods like co-pigmentation, dye engineering, anthocyanin dye structure modifications, and protective coatings. These avenues present promising opportunities to significantly enhance the efficiency, stability, and economic viability of anthocyanin-based DSSCs. This research contributes substantively to advancing sustainable solar energy technology, positioning anthocyanins as compelling and environmentally friendly alternatives in the realm of renewable energy. The findings underscore the potential of anthocyanin-based DSSCs as a viable solar energy technology, paving the way for further promising research and development in this field.

Keywords: anthocyanins, dye-sensitized solar cells, efficiency, stability, economic viability

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1. Introduction

Solar energy has emerged as a highly promising renewable energy source (Devadiga et al., 2021; Kokkonen et al., 2021), but the environmental impact of non-biodegradable substrates used in solar cell production poses a challenge for sustainable electronic devices (Gatto et al., 2022). In this context, the development of environmentally friendly and biodegradable materials is essential, particularly in dye-sensitized solar cells (DSSCs). DSSCs are cost-effective and easy to fabricate, and the use of natural dyes as sensitizers offers controlled modification of optical properties at a low cost, making them an attractive green alternative in renewable energy technology (Hosseinnezhad et al., 2020; Shalini et al., 2015).

In the Scopus database, there are 1,610 research papers discussing the use of natural dyes in solar cells. On average, 133 articles are published per year on this topic. Out of these articles, 1,431, 625, and 18 specifically address the subjects of efficiency, stability, and economy, respectively. Roughly 5.93% of the reviewed papers concentrate on efficiency, while approximately 11.52% discuss stability, and around 27.78% cover economic aspects. When the keyword search is filtered with terms like "anthocyanins", "betalains", "carotenoids", "chlorophyll", and "curcumin" the results show that these dyes make up 50%, 7%, 6%, 32%, and 5% of the papers, respectively. As Figure 1 illustrates, anthocyanins, chlorophyll, curcumin, carotenoids, and betalains are among the natural dyes. Among them, anthocyanins are the most commonly used natural dye in DSSCs, accounting for approximately 50% of their usage. Anthocyanins have demonstrated remarkable properties and have attracted significant attention for their potential applications in DSSCs.

The efficiency of anthocyanin-based DSSCs can vary depending on the specific dye and cell optimization. Earlier investigations have documented that DSSCs employing natural dyes such as anthocyanins have displayed conversion efficiencies in the range of 0.01% to 2.6% (Shalini et al., 2015; Teja et al., 2022). These efficiencies are comparatively lower when compared to metal complexes and organic dyes, which are frequently utilized in DSSCs and have achieved efficiencies ranging from 8% to 12% (Chen et al., 2019; Kakiage et al., 2015; Krawczak, 2019; Yamaguchi et al., 2010). The lower efficiencies of natural dye-based DSSCs can be attributed to the low molar extinction coefficients of natural dyes, resulting in reduced light absorption and photocurrent generation. Additionally, the broad absorption bands of natural dyes can limit electron injection into the TiO_2 semiconductor.

Efforts have been made to improve the efficiency of natural dye-based DSSCs through modifications in dye structure and co-sensitization techniques, such as blending, co-pigmentation, and acidifying of anthocyanin dyes, which can enhance the energy conversion efficiency of the cells. However, despite numerous studies and literature reviews, the efficiency of anthocyanin-based DSSCs still remains lower compared to chlorophyll-based and metal-complexed dye-based DSSCs. This suggests the need for more in-depth research to better understand the dye sensitization process and further improve the overall efficiency of anthocyanin-based DSSCs.

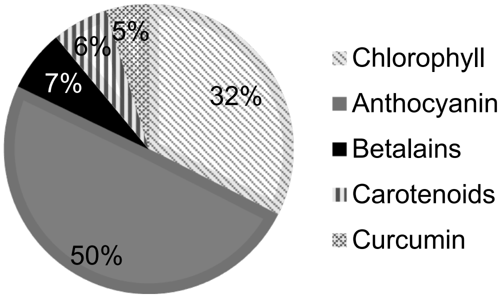

Figure 1. Anthocyanins, chlorophyll, curcumin, carotenoids, and betalains studied for DSSCs.

In addition to efficiency, natural anthocyanin-based dye-sensitized solar cells (DSSCs) face two other important challenges: stability and affordability. Stability is crucial for long-term performance, and factors such as pH, structure, temperature, and light can affect the stability of anthocyanin-based DSSCs (Amogne et al., 2020; Bhogaita et al., 2016; Câmara et al., 2022; Chawla & Tripathi, 2015; Kumara et al., 2017; Ludin et al., 2014; Shalini et al., 2015). Therefore, there is still a gap in the reviews that focus on understanding these influences and devising strategies to address stability issues. This includes exploring anthocyanin derivatives that are tolerant to pH variations, investigating methods to modify the dye structure for enhanced binding and electron transfer, and developing protective coatings for dyes. Meanwhile, regarding affordability, anthocyanins offer economic advantages as alternative colorants to expensive metal complex sensitizers. However, the scalability of anthocyanin extraction methods, market demand, and commercialization potential need to be reviewed to assess the economic viability of anthocyanin-based DSSCs.

Among the 1,610 papers, there are 13 paper reviews that discuss natural dye-based dye-sensitized solar cells (DSSCs) with a focus on anthocyanins, as shown in Table 1. Out of these, four paper reviews specifically tackle the topics of efficiency, stability, and affordability. One review explores the potential of peptides in DSSCs, emphasizing their unique properties and promising role as dyes and templating materials (Gatto et al., 2022). Another review investigates the efficiency of DSSCs by studying various dyes and their compatibility with different components, highlighting the potential of natural algal and bacterial dyes (Orona-Navar et al., 2021). A third review discusses molecular engineering, metal complexes, organic dyes, and natural dyes as photosensitizers, along with the environmentally friendly nature of solar cells (Giribabu et al., 2012). The fourth review evaluates the broader context of the energy transition to renewables, recognizing solar energy as an exploitable and adaptable source (Mitrašinović, 2021). Despite these reviews, a comprehensive assessment of the techno-economic factors and stability of DSSCs utilizing natural dyes, specifically anthocyanin dyes, is still lacking.

Table 1. A total of 13 paper reviews that discuss natural dye-based DSSCs, with a particular emphasis on anthocyanins as the natural dye.

No	Title	Natural dyes used	Main findings	Efficiency	Economy	Stability	Ref.
1	Behind the scenes of anthocyanins—from the health benefits to potential applications in food, pharmaceutical and cosmetic fields	Anthocyanins	-Anthocyanins have strong anti-inflammatory and antioxidant properties. -They are valuable in industries like textiles, food, pharmaceuticals, and cosmetics.	-	Economically viable alternative to conventional colorants	Stability affected by pH, structure, temperature, and light.	(Câmara et al., 2022)
2	Review on natural dye sensitized solar cells: Operation, materials, and methods	Natural dyes (including anthocyanin)	-Natural dyes can be used as sensitizers in DSSCs instead of synthetic dyes. -Natural dyes offer advantages such as affordability, biodegradability, and reduced environmental impact.	-	Low cost, biodegradable	Stability affected by extraction method, concentration, and pH sensitivity.	(Shalini et al., 2015)
3	Recent advances in anthocyanin dyes extracted from plants for dye sensitized solar cell	Anthocyanin	-Blending, co-pigmentation, and acidifying of anthocyanin dyes can improve of efficiency in DSSCs. -Solvent choice also affects stability.	-	Low cost, complete biodegradability	Stability affected by blending, co-pigmentation, and acidifying techniques.	(Amogne et al., 2020)
4	Recent advances in hybrid solar cells based on natural dye extracts from Indian plant pigment as sensitizers	Plant pigments (including anthocyanin)	-Natural dye extracts from plant pigments, such as anthocyanin, have been explored for use in hybrid solar cells. -Challenges include pH sensitivity, absorption in the near IR range, and electrolyte leakage.	-	Biodegradable, non-toxic	Stability affected by pH sensitivity, absorption properties, and electrolyte leakage.	(Bhogaita et al., 2016)
5	Novel improvements in the sensitizers of dye-sensitized solar cells for enhancement in efficiency-a review	Various sensitizers, including organic and natural dyes (e.g., anthocyanin)	-Organic and natural sensitizers offer potential for optimizing efficiency in DSSCs. -Natural sensitizers like anthocyanin and chlorophyll are cost-effective and environmentally friendly.	-	Cost-effective, environmentally friendly	Efficiency varies depending on the sensitizers used.	(Chawla & Tripathi, 2015)
6	Natural dye sensitized solar cells-a review	Natural dyes (e.g., anthocyanin, chlorophyll)	-Natural dyes from fruits, vegetables, leaves, flowers, and algae can be used as sensitizers in DSSCs. -Maximum conversion efficiencies achieved range from 0.01% to 2.6% depending on the dye used.	Up to 2.6% (depending on the dye)	-	-	(Sahni, 2012)

No	Title	Natural dyes used	Main findings	Efficiency	Economy	Stability	Ref.
7	Mapping the progress in natural dye-sensitized solar cells: Materials, parameters and durability	Anthocyanins, carotenoids, flavonoids, chlorophylls, tannins, betalains	Investigates natural sources of dyes, shortcomings, improvements in efficiency and stability, developments, and commercialization.	-	-	-	(Alim et al., 2022)
8	Review on the development of natural dye photosensitizer for dye-sensitized solar cells	Anthocyanin, carotenoid, chlorophyll, flavonoid	Natural dyes can cut down the high cost of noble metals and chemical synthesis in DSSCs.	-	economical due to the use of natural dyes.	Emphasizes the photostability of the sensitizer material.	(Ludin et al., 2014)
9	Review on the progress of light harvesting natural pigments as DSSC sensitizers with high potency	Natural sensitizers: chlorophyll, anthocyanin, carotenoid, flavonoid	Natural dyes can enhance photo-conversion efficiency in DSSCs.	High potency and durability	-	-	(Prakash & Janarthanan, 2023)
10	Natural dyes for dye sensitized solar cell: A review	Natural dyes: anthocyanin, carotenoid, flavonoid, chlorophyll pigments	Natural dyes can cut down the high cost of metal complex sensitizers and replace expensive chemical synthesis.	-	economical due to the use of natural dyes.	-	(Richhariya et al., 2017)
11	Progress in dye sensitized solar cell by incorporating natural photosensitizers	Natural dyes: chlorophyll, carotenoid, betalains, anthocyanin, flavonoid pigments	Natural dyes offer facile preparation, cost-effectiveness, and environmental friendliness in DSSCs.	Higher efficiencies and better stabilities	-	-	(Iqbal et al., 2019)
12	Review on fabrication methodologies and its impacts on performance of dye-sensitized solar cells	Natural dyes: betalains, anthocyanin	Natural dyes are economical, readily available, and environmentally friendly in DSSCs.	Efficiency improvements with optimized photoanodes and electrode doping	economical due to the use of natural dyes.	-	(Richhariya et al., 2022)
13	Recent progress and utilization of natural pigments in dye sensitized solar cells: A review	Anthocyanin, carotenoid, aurone, chlorophyll, tannin, betalains, and others	Natural dyes have high absorption coefficients, low-cost extraction, and low toxicity in DSSCs.	Lower efficiency compared to metal complexes and organic dyes.	economical due to the use of natural dyes.	Emphasizes the advantages and limitations of natural dyes.	(Kumara et al., 2017)

Dye-sensitized solar cell (DSSC) research has recently undergone a significant shift, emphasizing the use of anthocyanins as potent sensitizers. These natural pigments, abundant in fruits and plant tissues, are recognized for exceptional light-absorbing properties. Seminal previous works in 2023 highlight the efficacy of anthocyanins from diverse sources (Aziza et al., 2023; Montagni et al., 2023; Rajoriya et al., 2023; Shukor et al., 2023; Soosairaj et al., 2023; Yadav et al., 2023). The chemical structure of anthocyanins, with conjugated double bonds and hydroxyl groups, facilitates efficient light absorption and charge transfer in solar cells. Recent advancements showcase the potential of anthocyanins to enhance power conversion efficiencies in DSSCs (Adu et al., 2022; Al Batty et al., 2022; Mejica et al., 2022). This collective state-of-the-art, incorporating sixteen research works, positions anthocyanins as competitive alternatives to conventional sensitizers. Anthocyanins from various sources, including blackberries, acai berries, *Leucophyllum frutescens*, and Malabar spinach (*Basella alba*), have substantially improved power conversion efficiency, reaching up to 13.79 and 0.122 μW for outdoor and indoor environments, respectively (Shukor et al., 2023). Innovative approaches, like combining anthocyanins from different fruits demonstrate the versatility of anthocyanin-based DSSCs (Al Batty et al., 2022). This transformative phase in DSSC research positions anthocyanins as leading contenders for sustainable solar cell technologies, offering competitive efficiencies and potential applications in educational kits and technology development initiatives (Shukor et al., 2023). This collective research, spanning diverse sources and innovative approaches, underscores the promising trajectory of anthocyanins in advancing renewable energy.

This critical review aims to evaluate the current status of anthocyanin-based DSSCs, focusing on efficiency, stability, and economic viability. It analyses recent research developments, experimental techniques, and performance indicators to assess advancements and propose strategies for enhancing the efficiency and stability of these solar cells. Additionally, it explores economic considerations, availability, cost, scalability, and innovative approaches such as dye engineering and novel device architectures. By providing valuable insights, this study seeks to highlight the potential of anthocyanin-based DSSCs as a sustainable and economically feasible technology for solar energy harnessing.

2. Methods

In the pursuit of comprehensively evaluating the potential of anthocyanins as natural dyes in solar cells, the research methodology underwent a rigorous series of steps, each carefully calibrated and validated. The initial phase involved a meticulous calibration of search algorithms for databases like Scopus and Google Scholar. This calibration aimed to optimize the retrieval of pertinent articles related to anthocyanins, natural dyes, and solar cells. The subsequent validation of the search strategy ensured its effectiveness by comparing the retrieved articles against a predefined set of key articles and establishing benchmarks for relevance. Similarly, the process of keyword selection underwent systematic calibration and validation, refining the chosen keywords to enhance precision and relevance in retrieving literature. Inclusion criteria, pivotal to the study's focus, were subjected to careful calibration, defining, and fine-tuning to ensure consistency and relevance. The validation of these criteria was executed through a pilot test on a subset of articles, confirming their consistent application among researchers.

The meticulous design and refinement of data extraction forms constituted a calibrated effort aimed at consistently capturing relevant information throughout the research process. These forms underwent validation through pilot testing on a subset of articles, ensuring their effectiveness in extracting pertinent data. Likewise, the analytical methods employed for data analysis were subject to calibration and validation, guaranteeing their ability to identify trends and significant findings with both meaningfulness and reliability. The ongoing calibration and validation efforts were fundamental to maintaining the study's integrity, underscoring their integral role in the entire research endeavor.

As the research progressed, these systematic calibration and validation processes played a pivotal role in shaping the study's outcome. It culminated in a comprehensive summary and in-depth discussion of the research findings, providing a nuanced understanding of the subject matter. The critical evaluation of the reviewed studies, facilitated by meticulous calibration and validation, added depth to the analysis. Additionally, the research's suggestions for further exploration and study directions were informed by the robustness of the methodology. The sequential stages of this rigorous and validated review process are visually encapsulated in Figure 2, offering a transparent representation of the methodological rigor employed in this scholarly pursuit.

3. Results and discussion

3.1. Physical and chemical properties of anthocyanins for DSSCs

The examination of the physical and chemical properties of anthocyanins for dye-sensitized solar cells (DSSCs) reveals crucial factors influencing their performance. Key properties include strong light absorption within the visible range, alignment of energy levels with semiconductor materials, efficient electron injection, stability under light and heat exposure, solubility for extraction and deposition, suppression of charge recombination, and matching redox potential with the electrolyte.

Several studies on anthocyanin dyes demonstrate absorption peaks within the range of 400 to 600 nm as shown in Figure 3, showcasing their suitability for light absorption in DSSCs. Distinct peaks are noted at marginally varying wavelengths, including 520, 532, or 543 nm (Abdullah et al., 2022; Chang & Lo, 2010; Noor et al., 2014; Syafinar et al., 2015; Widhiyanuriyawan et al., 2022). Various origins of anthocyanins, such as mangosteen fruit peel, blueberries, and black rice, manifest distinctive absorption peaks in the visible light spectrum. To illustrate, anthocyanin dyes derived from mangosteen fruit peel display light absorption capabilities spanning from 480 to 580 nm (Widhiyanuriyawan et al., 2022), while blueberry anthocyanins showcase peak absorbance at 500 nm, encompassing a broader absorption range within the visible light spectrum (450–600 nm) (Syafinar et al., 2015). The anthocyanin isolated from black rice exhibits a distinct absorption peak at 532 nm (Noor et al., 2014). Additionally, studies reveal that xanthones and anthocyanins, often acquired concurrently in the extraction process, present separate absorption peaks at various wavelengths. Moreover, the pH of the environment affects anthocyanin structure and colors, impacting light absorption and electron generation (Pramananda et al., 2021).

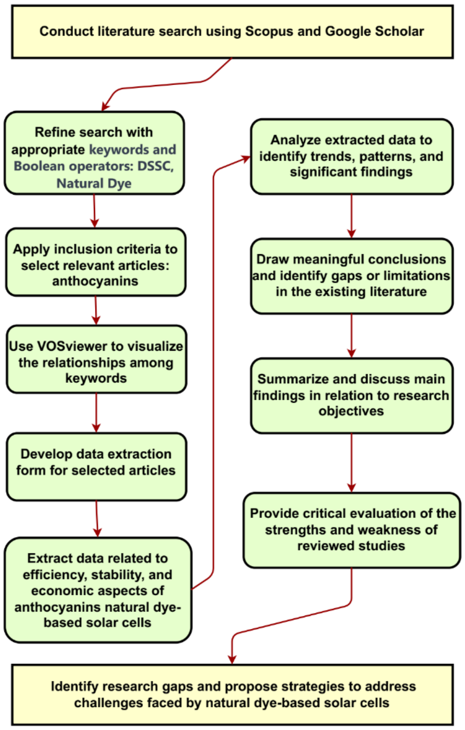

Figure 2. Stages involved in the comprehensive review process.

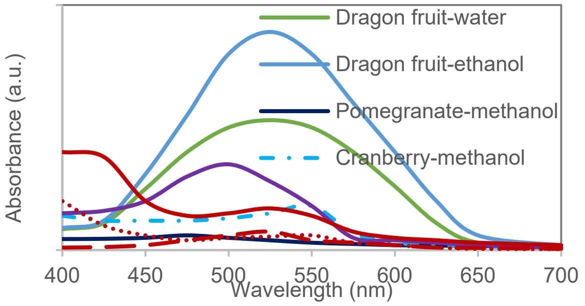

Figure 3. UV-visible spectra of some anthocyanin dyes (Abdullah et al., 2022; Chang & Lo, 2010; Noor et al., 2014; Syafinar et al., 2015; Widhiyanuriyawan et al., 2022).

These findings collectively emphasize that anthocyanin dyes possess absorption peaks falling within the 400 to 600 nm range, rendering them suitable for light absorption in DSSCs (Mulinacci et al., 2008). Furthermore, the unique absorption peaks displayed by anthocyanins from diverse sources open avenues for refining the light absorption properties of DSSCs. Researchers have the opportunity to explore anthocyanins with absorption peaks at specified wavelengths, incorporating them into sensitized electrodes to enhance the targeting of specific regions within the solar spectrum more effectively.

Furthermore, the shift towards longer wavelengths in absorption peaks when utilizing anthocyanin-sensitized TiO_2 films signifies altered absorption characteristics, leading to a more extensive light absorption spectrum compared to the sole dye solution (Nan et al., 2017). Anthocyanins, characterized by water-solubility and a broad absorption range owing to conjugated π bonds as shown in Figure 4, emerge as a promising avenue for augmenting DSSC efficiency by capturing a diverse sunlight spectrum (Pramananda et al., 2021). The observed redshift in absorption peaks implies that electrodes sensitized with anthocyanins can effectively capture a broader array of light, encompassing photons with lower energy levels. This enhancement contributes to a more holistic exploitation of solar energy, a critical factor for DSSCs reliant on proficient light absorption to generate electricity (Nan et al., 2017).

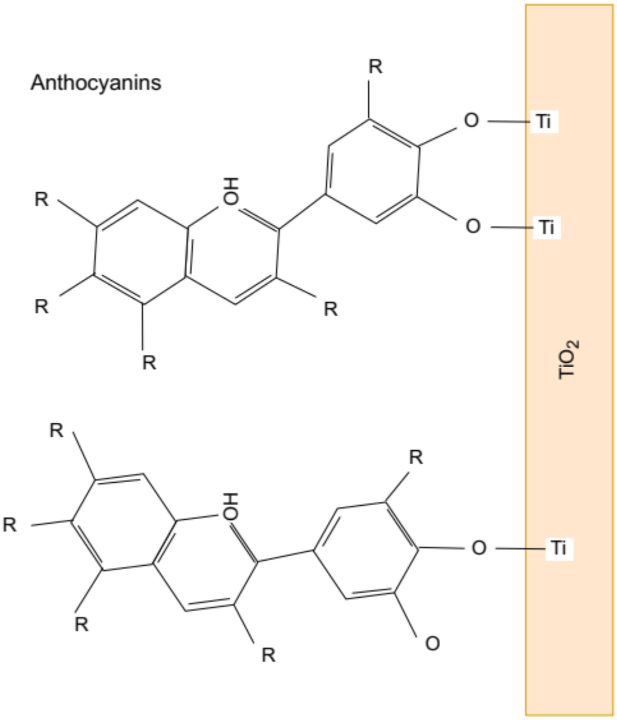

Figure. 4. Basic structure of anthocyanins attached onto TiO_2 (Nan et al., 2017; Pramananda et al., 2021).

The alignment of energy levels between anthocyanin dyes and semiconductor materials, as depicted in energy level diagrams in Figure 5, confirms the viability of efficient electron transfer and charge collection in DSSCs (Setiarso & Sova, 2023). Delphinidin, a specific anthocyanin, exhibits HOMO and LUMO values conducive to efficient electron transfer, emphasizing the importance of favorable energy level alignments (Ghann et al., 2017). Structural characteristics, including conjugation and specific molecular groups, further facilitate rapid electron movement and efficient injection into semiconductors (Ghann et al., 2017; Sreeja & Pesala, 2019).

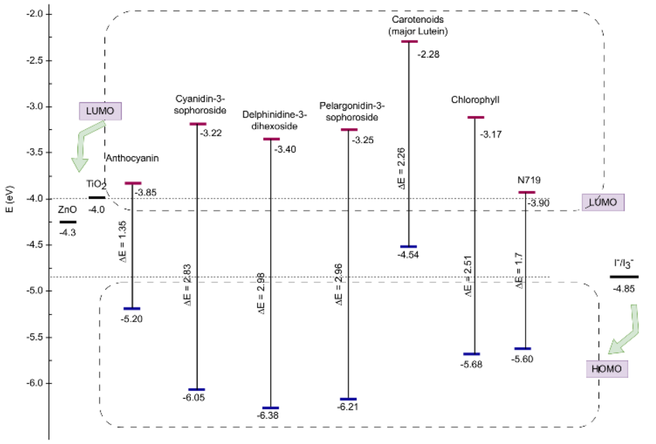

Figure 5. Energy level diagram of different natural dyes, N719, electrolyte, TiO_2, and ZnO (Ghann et al., 2017; Prabavathy, Shalini, Balasundaraprabhu, Velauthapillai, Prasanna, & Muthukumarasamy, 2017; Setiarso & Sova, 2023; Sreeja & Pesala, 2019).

To address the challenge of charge recombination in DSSCs, strategies such as molecular engineering, co-sensitization, interface engineering, and electrolyte optimization are employed (Setiarso & Sova, 2023). Modification of anthocyanin molecular structures enhances electron separation, co-sensitization broadens light absorption, and optimizing interfaces and electrolytes contributes to suppressing recombination, crucial for DSSC efficiency. Observations reveal a minimum energy gap of 0.2 eV for effective electron injection from anthocyanin dyes to the TiO_2 conduction band (Prabavathy, Shalini, Balasundaraprabhu, Velauthapillai, Prasanna, & Muthukumarasamy, 2017). Delphinidin, found in pomegranate fruit, exhibits HOMO and LUMO values promoting efficient electron transfer to TiO_2, with a reduced band gap enhancing intramolecular electronic transitions (Ghann et al., 2017). Energy level diagrams in Figure 5 illustrate the alignment of anthocyanin HOMO with electrolyte redox potential and E_{LUMO} with TiO_2 or ZnO energy levels, facilitating regeneration and electron injection (Setiarso & Sova, 2023).

Light interaction with anthocyanins in DSSCs results in absorbed light, altering energy levels and generating electrons for injection into TiO_2 or ZnO semiconductors (Nan et al., 2017). The energy difference of 0.15–0.19 eV between TiO_2 and anthocyanin signifies a high electron injection driving force, enhancing charge collection efficiency (Nan et al., 2017). Ultrasonic extraction of blueberry anthocyanins reveals a photon energy gap of 2.49 eV, indicating the required energy range for effective light absorption and electron excitation (Syafinar et al., 2015).

Efficient electron injection by anthocyanins in DSSCs relies on factors such as energy level alignment, functional groups facilitating metal complex formation, structural characteristics promoting electron delocalization, and optimized pH conditions (Buraidah et al., 2011; Mejica et al., 2022; Okello et al., 2022). Solubility of anthocyanins, influenced by pH, solvent polarity, and temperature, is crucial for extraction and deposition (Prakash & Janarthanan, 2023; Ramirez-Perez et al., 2019; Soonmin et al., 2023; Zulkifili et al., 2015). Strategies, including solubilizing agents, enhance solubility for uniform coating.

The redox potential alignment between anthocyanins and electrolytes is crucial for efficient electron transfer in DSSCs (Prajapat et al., 2023). The redox potential of the electrolyte must match that of the dye to facilitate effective electron transfer processes, complete the electron transfer cycle, and suppress charge recombination. Efforts to suppress charge recombination involve molecular engineering, co-sensitization, interface engineering, and electrolyte optimization (Prajapat et al., 2023). Modification of anthocyanin molecular structure, co-sensitization with complementary dyes, optimizing interfaces, and selecting suitable electrolyte compositions contribute to minimizing recombination losses (Prajapat et al., 2023).

The study on anthocyanins for dye-sensitized solar cells (DSSCs) reveals several strengths that contribute to the advancement of solar energy conversion technology. One notable strength lies in the diverse absorption peaks exhibited by anthocyanin dyes sourced from various origins. This diversity not only underscores the adaptability of anthocyanins but also provides researchers with a valuable opportunity to refine light absorption properties in DSSCs. By exploring anthocyanins with specific absorption wavelengths, there is potential to enhance the targeting of distinct regions within the solar spectrum, optimizing energy capture. Furthermore, the application of anthocyanins in sensitizing TiO_2 films results in an enhanced light absorption spectrum. The observed redshift in absorption peaks leads to altered absorption characteristics, expanding the range of light absorption compared to using sole dye solutions. This enhancement is crucial for DSSCs, which heavily rely on proficient light absorption for efficient electricity generation. Another notable strength lies in the study's focus on efficient electron transfer. The alignment of energy levels between anthocyanin dyes and semiconductor materials, exemplified by the case of delphinidin, signifies favorable energy level alignments for rapid electron movement and injection into semiconductors. This alignment aligns with the structural characteristics of anthocyanins, including conjugation and specific molecular groups, contributing to the overall efficiency of electron transfer processes in DSSCs. Additionally, the study addresses the challenge of charge recombination in DSSCs through various strategies such as molecular engineering, co-sensitization, interface engineering, and electrolyte optimization. These approaches play a crucial role in suppressing recombination, thereby improving the overall efficiency of DSSCs. The study also provides quantifiable information on the energy gap required for effective electron injection from anthocyanin dyes to the TiO_2 conduction band. This data serves as a foundation for understanding and optimizing the electron injection process, offering valuable insights into enhancing DSSC performance. Moreover, the determination of a photon energy gap for blueberry anthocyanins through ultrasonic extraction contributes to a quantitative understanding of the light absorption properties of anthocyanins, further enhancing the study's strengths.

Despite these strengths, the study acknowledges several limitations that warrant consideration. One notable limitation is the source dependency of anthocyanin absorption peaks, varying based on the origin of the anthocyanin. This source dependency may limit the generalizability of findings, necessitating specific considerations for different anthocyanin sources. The complexity of absorption peaks poses another challenge, as the intricate nature of these peaks may hinder the precise tailoring of light absorption properties. Further investigation is needed to better understand and optimize this complexity for practical application. The observed redshift in absorption peaks when using anthocyanin-sensitized TiO_2 films is noted, but the impact on other film properties and device performance is not extensively discussed. This lack of detailed exploration leaves room for further investigation to comprehensively understand the implications of the observed redshift. The study also mentions various strategies for suppressing charge recombination, but the specific efficacy and trade-offs of each strategy are not thoroughly explored. Additional research is necessary to determine the most effective combination of strategies for optimal DSSC performance. Furthermore, the study briefly touches on the importance of anthocyanin solubility for extraction and deposition, but specific strategies and their effectiveness are not extensively discussed. Further exploration of solubilizing agents and co-solvents is warranted to enhance the practical application of anthocyanins. Lastly, while the study emphasizes the importance of redox potential alignment between anthocyanins and electrolytes for efficient electron transfer, the specific challenges and strategies for achieving and maintaining this alignment are not deeply explored. Further insights into this aspect are needed to enhance the practical application of anthocyanins in DSSCs.

3.2. Sources of anthocyanins for DSSCs

In this study, potential sources of anthocyanins for dye-sensitized solar cells (DSSCs) were investigated, with a focus on their efficiency and stability. Pomegranate and mangosteen peel, along with various other plant materials, were examined for their anthocyanin content. Pomegranate, recognized for its vibrant red hue, proved to contain anthocyanins such as delphinidin, cyanidin, and pelargonidin. The application of pomegranate-derived anthocyanin dyes resulted in DSSC efficiencies ranging from 0.1% to 1.5%, showcasing improvements in TiO_2 film sensitization and broadening the absorption range of visible light. Similarly, mangosteen peel, with its content of xanthones and anthoanthocyanins, demonstrated light absorption in the range of 480 to 580 nm. The study further explored additional sources of anthocyanins, each exhibiting varying DSSC efficiencies ranging from below 0.1% to 3.16% (Behjat et al., 2019; Chang & Lo, 2010; Erande et al., 2021; George et al., 2020; Hemmatzadeh & Jamali, 2015; Hosseinnezhad et al., 2015; Hosseinpanahi et al., 2017; Khammee & Ramaraj, 2021; N. Kumara et al., 2013; Mejica et al., 2022; Nan et al., 2017; Noor et al., 2014; Simiyu et al., 2004; Widhiyanuriyawan et al., 2022).

A comparative analysis with other studies, summarized in Table 2, highlighted the diverse performance of DSSCs using different anthocyanin sources and sensitized electrodes. Variations in efficiency, short-circuit current density (J_{SC}), open-circuit voltage (V_{OC}), and fill factor (FF) were observed across these systems, ranging from 0.122% to 3.16%. The nuanced differences in specific parameters were attributed to the choice of anthocyanin source and the type of sensitized electrode, emphasizing the importance of a tailored approach in optimizing DSSC efficiency.

Addressing challenges inherent in DSSCs using anthocyanins, the study focused on improving the fill factor, enhancing current, and optimizing voltage. These challenges necessitated the refinement of light absorption efficiency, electron transfer efficiency, charge carrier recombination, and positive charge transfer efficiency. Emphasizing the need for further research and development, the findings underscored the importance of refining device design, materials, and manufacturing processes to enhance the overall efficiency of DSSCs as a sustainable solar energy alternative. The implications of the study extend beyond anthocyanins, providing insights into the broader field of renewable energy and emphasizing the necessity for a comprehensive approach to harnessing their potential. Acknowledging its strengths in identifying diverse anthocyanin sources and presenting a comprehensive performance summary in Table 2, the study also acknowledged limitations. These limitations include the source dependency of anthocyanin absorption peaks, the complexity of optimizing absorption properties, and the need for further exploration of solubility strategies. Challenges related to low fill factor, current enhancement, and voltage improvement were identified, indicating areas where future research efforts could contribute significantly to advancing DSSC technology. The study sets the groundwork for future investigations into anthocyanins and their role in improving solar energy utilization.

3.3. Strategies for enhancing efficiency of aanthocyanin-based DSSC

In exploring ways to enhance the performance and efficiency of dye-sensitized solar cells (DSSCs), this study investigates a range of methods, including the use of different dyes, co-pigmentation, pH optimization, dye mixing, sequential deposition, and hybrid dyes. The efficacy of these strategies becomes apparent when considering the broad spectrum of DSSC improvement opportunities. For instance, the examination of dye mixing demonstrates its capacity to expand the visible light absorption range and heighten electron excitation transmitted to TiO_2, a finding substantiated by various studies detailed in Table 2.

Comparisons with other studies underscore the significance of co-pigmentation, wherein anthocyanin interaction with co-pigment compounds, such as metal ions (Al^{3+}, Fe^{3+}, Sn^{2+}, Cu^{2+}) or organic compounds leads to increased photon absorption and enhanced efficiency. Studies exploring this interaction reveal the formation of complex and stable structures, contributing to overall improved efficiency. The study also delves into the pivotal role of pH optimization in influencing multiple parameters, showcasing the nuanced relationship between acidity, energy conversion, and power density. Lower pH conditions can lead to enhanced energy conversion efficiency, decreased bandgap, improved voltage at open circuit (V_{OC}) and short-circuit current density (J_{SC}), and slightly decreased fill factor (FF). For example, the power density of DSSCs increased as the pH decreased under acidic conditions, reaching its peak at pH 2. On the other hand, the Malabar spinach dye demonstrated optimal performance at alkaline pH 9, exhibiting the highest power density among the tested DSSCs. Moreover, the benefits of sequential deposition of dye onto the TiO_2 semiconductor are evident in the efficient electron injection into the semiconductor through anthocyanin carbonyl and hydroxyl groups. Additionally, hybrid dyes introduce a unique mechanism, distinct from co-sensitizer dyes, by having one dye act as a donor and transfer energy non-radiatively to the acceptor dye, enriching the overall efficiency of the solar cell.

Table 2. Performance of anthocyanin-based DSSCs.

Anthocyanins /Anode	J_{SC} (mA)	V_{OC} (mV)	FF	Efficiency (%)	Ref.
Bare TiO_2	0.99	300	0.22	0.17	(Erande et al., 2021)
Pomegranate/TiO_2	1.62	304	0.21	0.20	(Erande et al., 2021)
Pomegranate/TiO_2	3.1	420	0.46	0.61	(Behjat et al., 2019)
Mangosteen peel/TiO_2	0.517	506	0.48	0.126	(Widhiyanuriyawan et al., 2022)
Fruit of Malabar spinach/TiO_2	0.0682	487		0.102	(Mejica et al., 2022)
Troll flower/TiO_2	0.341	493	0.725	0.122	(Nan et al., 2017)
Mulberry/TiO_2	1.89	555	0.53	0.548	(Chang & Lo, 2010)
Eggplant in ethanol/cetone /TiO_2	1.57	430	0.86	0.58	(Hemmatzadeh & Jamali, 2015)
Eggplant in basified ethanol/cetone /TiO_2	2.62	470	0.74	0.91	(Hemmatzadeh & Jamali, 2015)
Eggplant in basified ethanol/cetone/distilled water (2:1:1)/TiO_2	3.81	540	0.39	0.80	(Hemmatzadeh & Jamali, 2015)
Red onion in basified ethanol/TiO_2	1.50	630	0.98	0.93	(Hemmatzadeh & Jamali, 2015)
Fruit sheath of *Nephelium lappaceum*	1.17	453	0.48	0.25	(N. Kumara et al., 2013)
Saffron petal/TiO_2	2.32	397	0.71	0.66	(Hosseinpanahi et al., 2017)
Kalanchoe flower/TiO_2	2.874	712	0.30	0.61	(George et al., 2020)
Anthocyanin extracted from *Melastoma malabathricum*'s flower petals/TiO_2	3.39	410	0.62	0.84	(Khammee & Ramaraj, 2021)
Anthocyanin extracted from sour grenade/TiO_2	0.50	297	0.49	0.73	(Hosseinnezhad et al., 2015)
Anthocyanin extracted from sweet grenade/TiO_2	0.62	460	0.55	1.57	(Hosseinnezhad et al., 2015)
Anthocyanin extracted from *Hibiscus sabdariffa*/TiO_2	27	230	0.51	3.16	(Simiyu et al., 2004)
Anthocyanin extracted from *Ribes nigra*/TiO_2	30	215	0.46	2.97	(Simiyu et al., 2004)
Turmeric + spinach + mangosteen (curcumin + chlorophyll + anthocyanin = 1:2:1)	2.174	471	0.50	0.513	(Widhiyanuriyawan et al., 2022)
(Mulberry + pomegranate leaf)/TiO_2	2.8	530	0.49	0.722	(Chang & Lo, 2010)
Black-rice and *Pandanus amaryllifolius* leaves	2.63	470	0.58	0.72	(Noor et al., 2014)

The implications of these findings are substantial for advancing DSSC technology. Dye mixing, by broadening the absorption range and enhancing electron excitation, holds promise for improving DSSC efficiency. Co-pigmentation, through the formation of stable structures, contributes to increased photon absorption, amplifying the overall efficiency of DSSCs. The nuanced role of pH optimization in influencing various parameters emphasizes the complexity of the relationship between acidity, energy conversion, and power density. Sequential deposition emerges as a strategic method, facilitating efficient electron injection and, consequently, enhancing DSSC efficiency. The unique mechanism of hybrid dyes, where one dye serves as a donor and transfers energy to the acceptor dye, opens promising avenues for enriching the overall efficiency of the solar cell.

The study's strength lies in its comprehensive exploration of various methods for enhancing DSSC efficiency. Incorporating studies demonstrating the effectiveness of dye mixing and co-pigmentation, coupled with insights into pH optimization, sequential deposition, and hybrid dyes, provides a holistic perspective. However, it is crucial to acknowledge the variability in effectiveness under specific experimental conditions and material contexts. This recognition highlights the need for further research and optimization to fully unlock the potential of these strategies in improving DSSC efficiency.

3.4. The stability of anthocyanins dye-based DSSCs

Anthocyanin stability in dye-sensitized solar cells (DSSCs) is a complex interplay of chemical and environmental factors. Chemical modifications, encompassing the addition of hydroxyl, methoxyl, glycosyl, and acyl groups, were identified as crucial for either enhancing or compromising stability. Simultaneously, environmental factors such as pH, temperature, light, and oxygen levels exerted significant influence (Amogne et al., 2020), as evidenced by the findings presented in Table 3. Extremes in pH, high temperatures, excessive light exposure (especially UV light), and oxygen presence were identified as contributors to the degradation and color changes in anthocyanins (Amogne et al., 2020; Roobha et al., 2011). This comprehensive understanding emphasizes the need for effective management of these factors to ensure the stability of anthocyanins, particularly in the context of their application in DSSCs.

Table 3. Mechanism of stabilization and factors affecting the anthocyanin stability.

No	Factors	Effect	Mechanisms of stabilization	Recommendation	Reference
1	pH	-pH affects the stability and color of anthocyanin pigments. -Anthocyanins are more stable in acidic medium (pH 4).	-Anthocyanins undergo structural transformations at different pH values, resulting in color changes. -At pH 4, the predominant form of anthocyanins is the flavylium ion, which is more stable.	-Maintain pH levels below 3 for red color, between 2 and 4 for blue color, and between 4 and 6 for coexistence of different forms. -Avoid pH values greater than 7 to prevent degradation. -Use monoglycosides and diglycoside derivatives, which are more stable under neutral pH conditions. -Adjust the pH of extracts to 4 using HCl or NaOH.	(Cabrita et al., 2000; Cooper-Driver, 2001; Fleschhut et al., 2006; Jackman et al., 1987; Vargas et al., 2013; Wahyuningsih et al., 2017)
2	Temperature	-Temperature influences the molecular structure of anthocyanins and accelerates their destruction. -Anthocyanins are less stable at higher temperatures (from 25 °C to 68 °C).	-Higher temperatures lead to increased destruction of anthocyanins. -Heat introduced during processing and storage accelerates chalones transformation and decomposition of anthocyanins.	-Store anthocyanin-containing products at lower temperatures to minimize color loss and degradation. -Avoid high temperatures during processing and storage. -Store the extracts at a temperature of 4 °C.	(Arrazola et al., 2014; Devi et al., 2012; Vargas et al., 2013; Yusoff et al., 2014)
3	Light	-Light exposure accelerates the destruction of anthocyanin pigments.	-Light exposure leads to increased destruction of anthocyanin pigments.	-Protect anthocyanin-containing products from light exposure to prevent color degradation. -Store in dark or opaque containers.	(Contreras-Lopez et al., 2014; Devi et al., 2012; Vargas et al., 2013)
4	Co-pigmentation	-Co-pigmentation reactions enhance and stabilize the color of anthocyanins.	-Co-pigments form interactions with anthocyanin pigments, blocking hydration and stabilizing their color. -Co-pigments stabilize anthocyanins through the hyperchromic effect or bathochromic shift in the absorption spectra. -Co-pigments bind with electron-deficient forms of anthocyanins through delocalized π-electron systems. -Different compounds can act as co-pigments, including flavonoids, alkaloids, amino acids, organic acids, nucleotides, polysaccharides, metallic ions, and even anthocyanins themselves (self-association).	-Explore the use of co-pigments to enhance and stabilize the color of anthocyanins. -Investigate the interaction systems and specific co-pigments to optimize stabilization.	(Amogne et al., 2020; Calogero et al., 2015; Cortez et al., 2017)

In the exploration of anthocyanin extraction, a study (Prabavathy, Shalini, Balasundaraprabhu, Velauthapillai, Prasanna, Walke, et al., 2017) highlighted citric acid as the most effective solvent, enhancing stability and promoting sensitization properties when combined with TiO_2 nanorods. DSSCs utilizing anthocyanins extracted with citric acid exhibited superior photovoltaic performance, achieving an efficiency of 0.83%. Conversely, ethanol, a commonly used but less stable solvent, showed reduced efficiency due to TiO_2-induced degradation. The study underscored the impact of pH on anthocyanin stability, with lower pH favoring stability and storage. Notably, citric acid, known for its safety and low corrosiveness, interacted with anthocyanins, stabilizing their color and inhibiting degradation by polyphenol oxidase (PPO), establishing it as a promising solvent choice.

Sunlight exposure emerged as a critical factor triggering photochemical reactions in anthocyanins, leading to the formation of reactive species and free radicals. This degradation negatively affected the light-absorbing capacity and overall performance of DSSCs. Compared to synthetic dyes RR and coumarin, anthocyanin demonstrated higher photostability, with the lowest degradation rate under UV-visible light exposure (Abdou et al., 2013). Furthermore, dragon fruit extracts exhibited optimal pigment retention under specific storage conditions (pH 4, 4˚ C, and darkness) emphasizing the potential of anthocyanin as a stable sensitizer for DSSCs (Vargas et al., 2013).

Heat exposure, another environmental factor, was identified as a significant influencer on anthocyanin stability. Elevated temperatures in the range of 60 to 80 °C accelerated degradation processes, inducing structural changes and reducing light-absorbing properties and electron injection efficiency (Modesto Junior et al., 2023). The study highlighted the importance of considering temperature conditions in maintaining anthocyanin stability in DSSCs.

Various strategies were proposed to enhance anthocyanin stability, ranging from adjusting acidic pH to 3–4 using HCl or NaOH (Cabrita et al., 2000; Cooper-Driver, 2001; Fleschhut et al., 2006; Jackman et al., 1987; Vargas et al., 2013; Wahyuningsih et al., 2017) and adding fructooligosaccharides (Escher et al., 2020) to synthesize anthocyanin hybrid nano pigments. These nano pigments, integrated into polyester-based bionanocomposites, offered a diverse spectrum of colors (Cunha et al., 2023; Micó-Vicent et al., 2021). Other factors such as ring substituents, hydroxyl or methoxyl groups in the B ring, acylation, and glycoside derivatives were identified as potential stability influencers (Enaru et al., 2021). Co-pigmentation with metal ions like Fe^{3+} and Al^{3+} showed promise in modifying the absorption spectrum of anthocyanins, improving both stability and energy levels (Maylinda et al., 2019).

Therefore, the study elucidates the intricate factors influencing anthocyanin stability in DSSCs and proposes practical strategies for improvement. The findings underscore the potential of anthocyanins as stable sensitizers for solar cells, with implications for sustainable and efficient energy technologies. Future research should delve deeper into understanding the mechanisms of anthocyanin stability, refining the proposed strategies, and exploring their long-term applications in renewable energy technologies.

3.5. The economy of anthocyanins dye-based DSSCs

Dye-sensitized solar cells (DSSCs) employing anthocyanin as the dye hold promise for building-integrated photovoltaic applications due to their favorable attributes, including responsiveness to low and diffused light, color tunability, and transparency (Calogero et al., 2015; Cornaro & Andreotti, 2013; Kumara et al., 2017). However, the transition from small laboratory cells to larger building-integrated PV modules introduces challenges, resulting in reduced cell performance, increased series resistances, and decreased fill factor (Calogero et al., 2015; Fakharuddin et al., 2013). Natural dyes, including anthocyanin, face hurdles such as lower efficiencies and module stability compared to metal complex dyes like Ru-based dyes (Calogero et al., 2015). Despite this, the cost advantage offered by natural dyes, including anthocyanin, over synthetic organic dyes makes them economically appealing (Calogero et al., 2015).

Economically, the long-term stability of natural dyes in DSSCs poses a significant challenge for commercial development, impacting the economic viability of solar cells. The lifespan of solar cells greatly influences the overall cost of ownership, including raw materials, manufacturing processes, and long-term stability. The cost of the dye constitutes a significant portion of the overall module cost, followed by TiO_2 (Kalowekamo & Baker, 2009). The photoanode, consisting of dye, TiO_2, and TCO, accounts for over 50% of the total manufacturing cost (Calogero et al., 2015). Cost reduction strategies involve quantifying costs to identify areas for optimization and making sustainable DSSCs more economically viable. Comparisons with other renewable energy technologies consider manufacturing costs, material availability, scalability, and energy conversion efficiency. The cost structure analysis of DSSCs reveals that the dye, TiO_2, and photoanode components significantly contribute to the overall manufacturing cost (Calogero et al., 2015). Anthocyanin-based DSSCs exhibit the highest cost per Wp within natural dye options, emphasizing the need for further cost-reduction efforts (Calogero et al., 2015).

To address economic challenges, strategies for cost reduction and commercialization focus on enhancing device stability and optimizing formulations, sealing techniques, and module engineering design. Companies such as Toyota-Asia, TDK, Fujikura, and Sony are actively engaged in commercializing DSSCs, primarily employing metal complex dyes (Bosio & Alessandro, 2011; Fakharuddin et al., 2013). However, the cost advantage of natural dyes, including anthocyanin, presents an opportunity in terms of the performance/price ratio (Calogero et al., 2015). Further research and development, coupled with optimization strategies, can pave the way for the commercialization of DSSCs based on natural dyes, including anthocyanin, in building-integrated PV applications. Take into account elements like income generation, expenses associated with acquiring customers, costs related to maintenance, and overall profitability. By measuring these factors, it becomes feasible to evaluate the financial viability and possible returns of this business model.

Efficiency enhancement and addressing manufacturing costs are critical considerations for anthocyanin-based DSSCs. Anthocyanin-based DSSCs exhibit lower short-circuit current density (J_{SC}) compared to synthetic-dye DSSCs, emphasizing the need for efficiency improvement for commercial viability (Simiyu et al., 2004). Manufacturing costs are affected by dyes, counter electrodes, and substrates, collectively contributing to 50%–60% of the total cost (Hashmi et al., 2011). Comparatively, advancements in silicon-based solar cells have significantly reduced production costs, necessitating continuous competitiveness improvements for DSSCs. The cost per watt has significantly declined from around $4 in U.S. dollars to approximately $0.66 in U.S. dollars (Hardin et al., 2012). Obstacles like the expenses associated with the sealing process, constituting 27% of the total cost (Hashmi et al., 2011), and the need for material replacements present potential risks of efficiency decline. This underscores the delicate equilibrium necessary for achieving cost-effective performance.

While the cost of anthocyanins is low (0.24 €/g), the price per Wp (watt-peak) for anthocyanin-based DSSCs (10 €/m^2) is higher compared to both organic (2.7 €/m^2) and Ru-based DSSCs (4 €/m^2), primarily due to lower anthocyanin concentration on TiO_2 and lower efficiency (Calogero et al., 2015). This is primarily due to the lower concentration of anthocyanins on TiO2 (5×10^{-5} mol/cm^{-3}) and lower efficiency (η) at 1% (Calogero et al., 2015). Therefore, with current technology where the efficiency of anthocyanin-based DSSCs is approaching 3.16% (Simiyu et al., 2004), the price per Wp for anthocyanin-based DSSCs could be around (3.2 €/m^2), which falls between the price per Wp of organic-based and Ru-based DSSCs. A performance-based business model integrating DSSCs into buildings, providing green energy as a service, presents a circular economy approach. Quantitative evaluations of the environmental consequences associated with the entire lifespan of DSSCs are essential, considering raw material extraction, manufacturing, energy consumption, and end-of-life management. Identifying stages with the greatest environmental impact enables the development of strategies to mitigate the overall carbon footprint of DSSCs.

Moreover, it is estimated the cost of traditional sandwich-structured DSSCs to be in the range of $90–$116 in U.S. dollars per square meter (m^2) based on projected future material costs (Kroon et al., 2007). To achieve a cost of $0.66 in U.S. dollars per W comparable to silicon solar cells, DSSCs would require module efficiencies in the range of 13.6%–17.6%. However, the highest reported efficiency for anthocyanin-based DSSCs to date is 3.16% (Simiyu et al., 2004). From the performance analysis in the previous subsection, it was found that the V_{OC} (open-circuit voltage) and FF (fill factor) of anthocyanin-based DSSCs reached 712 and 0.71 mV, respectively, which are comparable to the V_{OC} and FF of synthetic-based DSSCs. Therefore, a critical step towards achieving commercial viability for anthocyanin-based DSSCs is to enhance their efficiency performance, particularly by addressing the significantly lower J_{SC} (short-circuit current density) compared to synthetic-dye DSSCs.

Therefore, addressing the economic challenges and improving the efficiency of anthocyanin-based DSSCs are crucial for their widespread adoption as a cost-competitive alternative in building-integrated photovoltaic applications. Continuous research and development efforts are needed to enhance efficiency, reduce manufacturing costs, and realize the commercial potential of DSSCs utilizing anthocyanin as a sustainable and economically viable option in the renewable energy landscape.

4. Conclusions

This comprehensive review underscores the potential of anthocyanin-based dye-sensitized solar cells (DSSCs), showcasing absorption properties within the 400 to 600 nm range. Strategies like dye mixing and pH optimization offer diverse enhancement pathways, addressing challenges in stability and manufacturing costs. Anthocyanin-based DSSCs, despite a lower current density than synthetic counterparts, present cost advantages with a performance/price ratio. The study quantifies the energy gap for effective electron injection, indicating potential for optimization. Anthocyanin-based DSSCs exhibit 3.16% efficiency, emphasizing the need for improvements to reach commercial viability. The article contributes quantifiable information on charge recombination suppression, laying the foundation for advancements. Despite limitations in source-dependent absorption peaks, the study advances understanding, offering a comprehensive overview. The findings provide a foundational framework for future research, encouraging a holistic approach toward sustainable and economically viable DSSCs in the renewable energy landscape. General comments applaud the thorough examination, clarity, and critical insights, promoting further refinement in the field. Overall, this review significantly advances anthocyanin-based DSSCs, offering valuable data and insights for their integration into the renewable energy sector.

Conflict of interest

The authors declare that there is no conflict of interest.

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