Asian Journal of Chemistry; Vol. 38, No. 5 (2026), 1135-1141
ASIAN JOURNAL OF CHEMISTRY
https://doi.org/10.14233/ajchem.2026.35414

Enhanced Light Harvesting in Dye-Sensitised Solar Cells using Chlorophyll-Curcumin-Anthocyanin Natural Dye Blend

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Received: 6 December 2025 *Accepted*: 4 March 2026 *Published online*: 4 May 2026 AJC-22337

Natural dyes offer a promising alternative to synthetic dyes in dye-sensitised solar cells (DSSCs), though their narrow optical absorption limits the efficiency of DSSCs. In present study, five flower samples, five leaf samples, turmeric rhizomes and *Basella rubra* berries were chosen for initial screening based on their optical absorption characteristics. Following preliminary analysis of visible-region absorption, leaf samples, turmeric rhizomes and *Basella rubra* berries were selected for further investigation. Chlorophyll, curcumin and anthocyanin pigments were extracted from curry leaves, turmeric rhizomes and *Basella rubra* berries, respectively. Optical absorption properties of each dye were studied using UV-Visible absorption spectroscopy. The chlorophyll pigment exhibited prominent absorption peaks at 434 nm and 664 nm, while curcumin and anthocyanin showed absorption at 420 nm and 534 nm, respectively. Given that individual dyes absorb only within a specific wavelength range, a co-sensitisation method was adopted to enable the dye to absorb the broad region of the solar spectrum. The bi-pigment blend of chlorophyll and curcumin and the tri-pigment blend of chlorophyll, curcumin and anthocyanin, demonstrated an increase of 159% and 171% respectively, in absorption coefficient, relative to the individual dyes. This enhanced spectral response suggests potential for higher photocurrent density for DSSCs.

Keywords: Chlorophyll, Curcumin, Anthocyanin, Co-sensitisation, Tri-pigment blend dye, Dye-sensitised solar cells.

INTRODUCTION

The global energy demand has increased due to the immense population growth, urbanisation, industrialisation, climate change and transport growth [1-3]. Since antiquity, most of these requirements have been entrusted to non-renewable energy sources; the use of such sources harms the environment and consequently affects humans and other living organisms [4,5]. This highlights the importance of renewable energy [6]. Solar energy technologies play a significant role in human development. Among various solar cell technologies, dye-sensitised solar cells are considered simple and cost-effective [7]. In these systems, photoelectrochemical processes within the dye enable the conversion of light energy into electrical energy [8].

The criteria for selecting a dye are important; dyes with broader absorption in the visible and near-infrared regions of solar radiation are more suitable for DSSCs. Ruthenium-based synthetic dyes, which satisfy these conditions, are used in DSSCs [9], achieving a power conversion efficiency of 11-12% [10-12]. However, the presence of platinum group metals as ruthenium based dyes increases their cost and toxicity, posing environmental and health risks [13]. Alternatively, natural dyes, which are readily available, abundant, eco-friendly and cost-effective, are being explored. The major classes of natural dyes used as active materials in DSSCs include anthocyanins, betalains, carotenoids, chlorophylls and curcumin. Extensive studies have been conducted on DSSCs using natural dyes. For instance, Abdel-Latif *et al.* [14] extracted 11 dyes from three different trees using flowers, leaves, bark and roots. The dye extracted from leaves exhibited a maximum conversion efficiency of 0.4%, likely due to chlorophyll, whereas dyes from roots and bark showed lower efficiencies. Pratiwi *et al.* [15] fabricated DSSCs from anthocyanin dyes extracted from various sources, such as cabbage, mangosteen peel and dragon fruit, with efficiencies of 0.054%, 0.042% and 0.024%, respectively. They used distilled water for the extraction. Mejica *et al.* [16] extracted anthocyanin from Malabar spinach berries and red cabbage using a solvent composed of 0.4 M acetic acid-sodium acetate buffer at pH 1.0 and (0.1 M) HCl acid-KCl buffer at pH 4.5, achieving efficiencies of 0.16654% and 0.00231%, respectively. DSSCs fabricated with curcumin dye [17], extracted in ethanol and using ZnO as the photoanode, showed a conver-

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sion efficiency of 0.266%. In all these studies, DSSCs were fabricated using single dyes. A mixture of two dyes could potentially broaden the absorption and improve the efficiency of DSSCs. For example, a beetroot and green spinach blend dye in a 50%:50% ratio in a DSSC showed an efficiency of 0.601%, compared to the individual dye efficiencies of 0.562% and 0.047%, respectively [18]. DSSCs made from curcumin extracted from turmeric rhizomes and anthocyanin extracted from red spinach exhibited efficiencies of 0.38% and 0.13%, respectively. A blend dye comprising 60% curcumin and 40% anthocyanin achieved an efficiency of 1.078%, surpassing the performance of single dyes [19]. Moreover, a green and red spinach blend dye in a 20%:80% ratio demonstrated higher efficiency than individual dyes [20].

Although several studies have continuously reporting the improved performance of DSSCs using single natural dyes and binary dye blends, the absorption coverage remains limited due to incomplete spectral complementarity. Most bi-pigment systems enhance absorption only within restricted wavelength regions, leaving gaps in the visible spectrum, thereby limiting the overall photocurrent generation in DSSCs [7,21]. Extending co-sensitisation to a tri-pigment dye blend yields panchromatic light harvesting by combining dyes with absorption characteristics in complementary spectral regions. Such a system can enhance photon utilisation, improve spectral overlap with the solar spectrum and potentially increase photocurrent density beyond binary dye systems [22,23]. However, systematic optical investigations of tri-pigment natural dye blends remain scarce. The present work addresses this gap by developing a chlorophyll-curcumin-anthocyanin tri-pigment dye blend for enhanced light harvesting. The optical properties of dyes extracted from selected five flowers *viz. Rosa indica* (rose), *Nelumbo nucifera* (lotus), *Hibiscus rosa-sinensis* (hibiscus), *Nymphaea pubescens* (lily) and *Pentas lanceolata* (pentas), along with leaves of *Zingiber officinale* (ginger), *Murraya koenigii* (curry leaf), *Coccinia grandis* (ivy gourd), *Piper nigrum* (black pepper), *Basella rubra* (Malabar spinach) and rhizomes of curcuma longa (turmeric) and *Basella rubra* (Malabar spinach) berries. Dye samples were selected for co-sensitisation, based on their absorption bands in the visible region, aiming to enhance the dye absorption. The broader absorption of 171% of the tri-pigment blend enhances photon capture and device efficiency.

EXPERIMENTAL

Chemical used for the extraction of dye was purchased from Himedia. Ethanol was chosen as a universal solvent for all dye extractions from all the raw materials due to its low toxicity, good solubility for chlorophyll, curcumin and anthocyanins and its ability to provide a common solvent environment for comparative optical studies. Although acidic solvents are often used to stabilise anthocyanins, but ethanol was preferred to maintain uniform extraction conditions as well as to avoid chemical modification of chlorophyll [24] and curcumin, which could further influence co-sensitisation behaviour.

Sample collection and extraction: All the raw materials were collected from the locality of Mavelikkara, Kerala, India. Fig. 1 shows all the samples used for the study. Samples were cleaned under running water to remove any dust or dirt.

Fig. 1. Samples for dye extraction (a) flowers (b) leaves (c) turmeric (d) Malabar spinach (*Basella rubra*) berry

For preparing dye from flowers, only the petals were utilised. The separated petals were washed with distilled water, air-dried at room temperature for 1 h and cut into small pieces. Each flower sample (5 g) were immersed in 20 mL of ethanol and left undisturbed for 18 h at room temperature in the dark. The same procedure was followed for extracting dye from leaves as well. The extraction of dye from Malabar spinach berries involved first selecting properly ripened berries. Cleaning and drying procedures were followed as described earlier. Berries (5 g) were lightly crushed to separate the seeds. To the remaining pulp, 20 mL of ethanol was added and the mixture was sonicated for 10 min, then kept undisturbed for 18 h at room temperature in the dark. Fresh and cleaned turmeric was selected for curcumin extraction. Selected turmeric was cut into small pieces and allowed to dry completely for 4 weeks. Dried turmeric samples were ground using a mortar and pestle. Five grams of this powder were immersed in 20 mL of ethanol. The extracts of all the above-mentioned dyes were filtered through Whatman No. 1 filter paper and stored in dark to prevent degradation.

Preparation of single and blend dye solutions: All single dyes were prepared using 400 μL of extracted dye per 20 mL of ethanol. The prepared dyes are used for co-sensitisation of dyes that absorb in different regions of the visible spectrum. The blend dyes were prepared by mixing 100 μL of leaf extract (chlorophyll), 100 μL of curcumin extract and 400 μL of *B. rubra* berry extract (anthocyanin). The dye mixing ratio was fixed at 1:1:4 to balance the higher absorption intensity of anthocyanin with the other dyes.

Characterisation: UV-Vis absorption spectra of the extracted dyes were recorded using a Systronics Type 117 spectrophotometer (200-1100 nm), with measurements carried out in the 250-800 nm range for both individual and blended dyes. To ensure reproducibility, dye extractions were performed independently and spectra were recorded multiple times, with consistent peak positions and relative intensities observed within experimental uncertainty.

RESULTS AND DISCUSSION

Selection of natural extract

Absorption spectra analysis of single dyes extracted from flowers: The absorption spectra of all flower dyes are illustrated in Fig. 2. The dye extracted from lotus shows prominent absorption at 389 nm and a comparatively weaker absorption in the visible region at 533 nm. Pink rose absorbs only at one UV wavelength, 355 nm, while pentas shows absorption at two regions: 354 nm and 535 nm. Hibiscus absorbs both UV and visible regions, with higher absorption in the UV region at 280 and 343 nm and a weak absorption at 540 nm. Among these, lily and rose do not show any absorption in the visible region. Table-1 presents the maximum absorption wavelengths and the corresponding absorption intensities for each dye. Although floral pigments such as anthocyanins contribute colour, their limited visible absorption and instability make them unsuitable for DSSCs.

Absorption spectra analysis of single dyes extracted from leaves: Fig. 3a-e illustrate the UV-Visible absorption spectra of pigments extracted from all the leaf samples, studied in the 250-800 nm region. The pigment extracted from each of the leaf samples exhibit absorption peaks between 230-285 nm [25] and 324-334 nm [26] are attributed to flavonoids and their derivatives. Every pigment shows a prominent absorp-

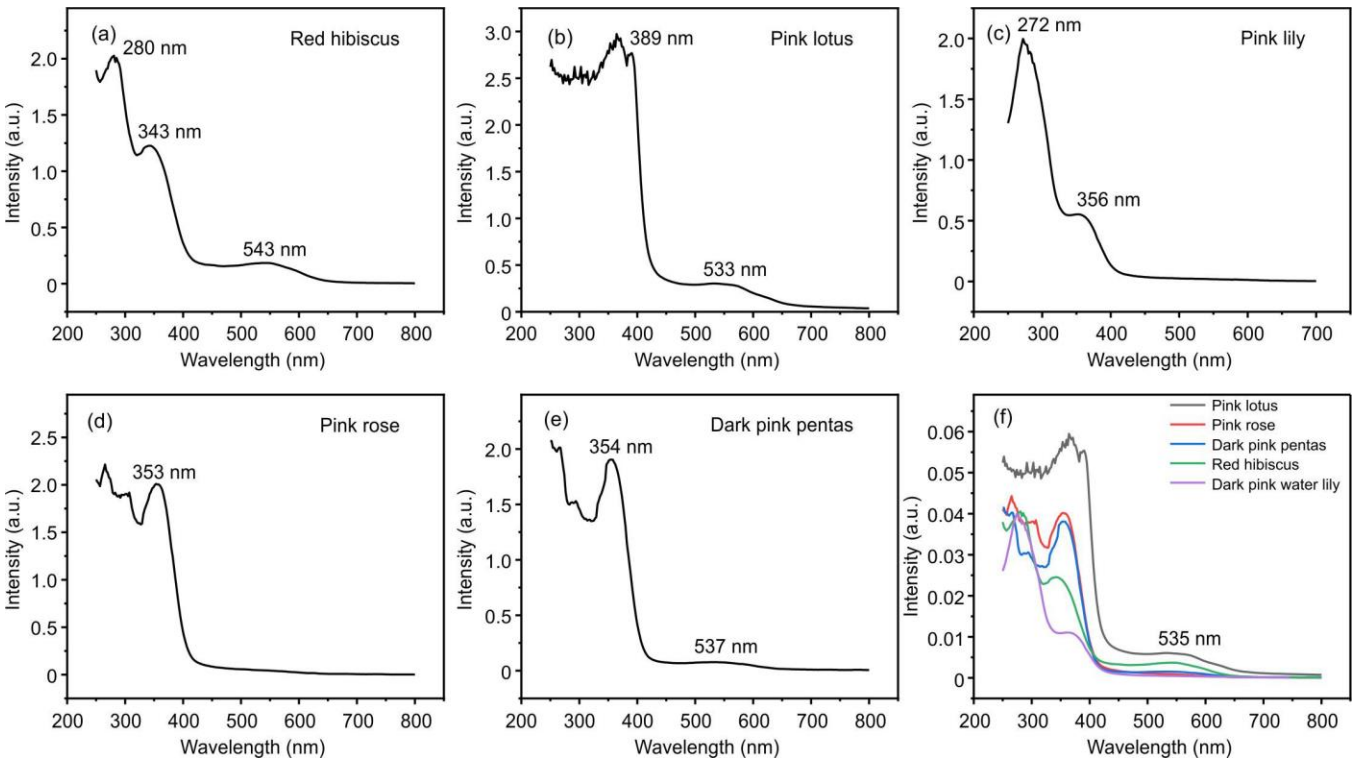

Fig. 2. Absorption spectra of hibiscus, lotus, lily, rose, pentas (a-e) and comparison of the absorption plots of all the flower extracts (f)

TABLE-1
MAXIMUM ABSORPTION WAVELENGTH AND CORRESPONDING INTENSITY OF STUDIED FLOWER DYES

Sample	UV region		Visible region	
	Max. absorption (nm)	Intensity (nm)	Max. absorption (nm)	Intensity (nm)
Pink lotus	389	0.05	533	0.007
Pink rose	355	0.04	–	
Dark pink pentas	354	0.04	535	0.003
Hibiscus	280, 343	0.04, 0.03	540	0.004
Dark pink lily	273, 368	0.04, 0.01	–	

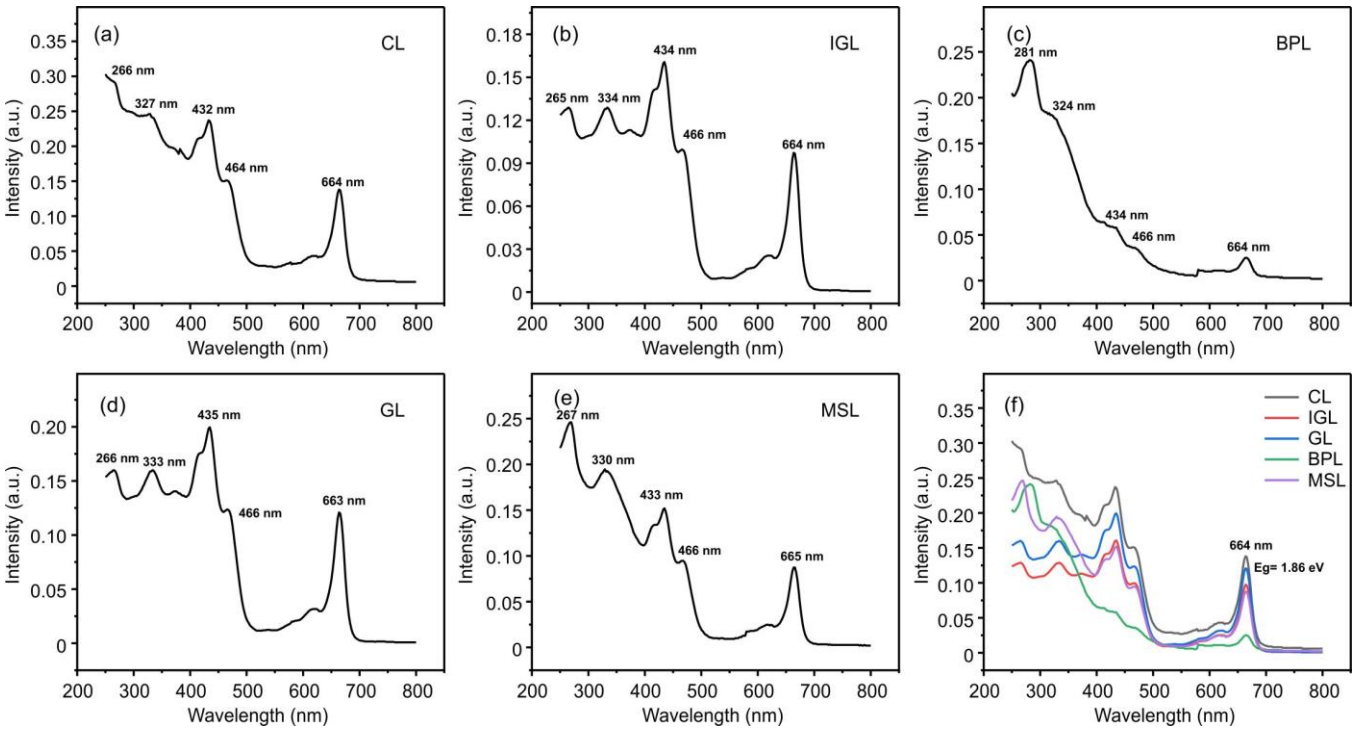

Fig. 3. Absorption spectra of pigment extracted from (a) curry leaf (CL), (b) ivy guard leaf (IGL), (c) black pepper leaf (BPL), (d) ginger leaf (GL), (e) Malabar spinach leaf (MSL) and (f) comparison of the absorption plots of all the leaf extracts

tion in 432-435 nm and at 664 nm, indicating the presence of the Soret band and Q band [27], respectively, confirming the presence of chlorophyll. The peaks at 464-466 nm are attributed to carotenoids [28], which act as the bridge between the Soret band and the Q band for charge transport [29]. Although absorption peaks of all the leaf extracts show the absorption in the visible region, the pigment extracted from curry leaf demonstrates better absorption in this region, as shown in Fig. 3f and Table-2. Therefore, it can be utilised in DSSC as a photosensitiser, either alone or co-sensitised with other pigments to enhance photon absorption.

Absorption spectra analysis of dye blends: One of the key factors in enhancing the efficiency of DSSC is the absorption of light by the dye. The selected photosensitiser dye must absorb a broad range of the visible spectrum of sunlight. From the previous section, it is evident that owing to limited absorption of visible light by flowers, they are not apt to proceed further for co-sensitisation, whereas leaf extracts are highly effective absorbers, absorbing at specific wavelengths (328, 433, 463 and 663 nm). The curcumin (~423 nm) and *B. rubra* berry (~534 nm) pigments absorb particular wavelengths in the visible region. Co-sensitisation was utilised to

TABLE-2
MAXIMUM ABSORPTION WAVELENGTH AND CORRESPONDING INTENSITY VALUES OF LEAF DYES

Absorption region / Samples	Intensity		
	Chlorophyll a (Soret band ~434 nm)	Carotenoid (~465 nm)	Chlorophyll a (Q band ~664 nm)
Curry leaf (CL)	1.30	0.76	0.70
Ivy guard leaf (IGL)	0.05	0.03	0.02
Ginger leaf (GL)	0.50	0.30	0.30
Black pepper leaf (BPL)	0.22	0.14	0.13
Malabar spinach leaf (MSL)	0.23	0.15	0.13

enhance absorption. The curry leaf extract was combined with curcumin and *B. rubra* berry (BRB) extracts. Fig. 4a-c show the absorption spectra of curry leaf, curcumin and BRB, while 4d and 4e illustrate the absorption of bi-pigment and tri-pigment extract, respectively. The concentrations of single dyes selected for co-sensitisation were estimated semi-quantitatively using Beer–Lambert law:

$$c = \frac{A}{\varepsilon l} \quad (1)$$

where c = the concentration of the dye, A = absorbance corresponding to λ_{max}, ε = molar extinction coefficient, l = path length (1 cm).

The absorbance values at the characteristic wavelength maxima of chlorophyll (~664 nm), curcumin (~423 nm) and anthocyanin (~534 nm) were used along with literature-reported molar extinction coefficients. Table-3 shows the concentrations of single dyes used for co-sensitisation.

TABLE-3
ABSORBANCE VALUES AT CHARACTERISTIC WAVELENGTH MAXIMA (λ_{max}), MOLAR EXTINCTION COEFFICIENTS (ε) AND CALCULATED CONCENTRATIONS OF INDIVIDUAL DYES

Dye	Absorbance corresponding to λ_{max}	ε (L mol^{-1} cm^{-1})	Concentration c = A/εl
Chlorophyll a	0.125	~8.7×10^4	1.463×10^{-6}
Curcumin	0.790	~5.5×10^4	1.463×10^{-5}
Anthocyanin	0.540	~2.6×10^4	2.070×10^{-5}

These concentration values represent approximate estimates intended for comparative analysis of single dyes and dye blends rather than absolute quantification. Upon co-sensitisation increase in absorption due to dye mixing was analysed by examining the area under each absorption curve and the absorption coefficient (α). Fig. 5 shows the absorption curves of single, bi-pigment and tri-pigment dye blends. The absolute area was calculated based on the integrated area under the absorption curves over the 250-800 nm wavelength range, using Origin Software. The absolute area under each curve was determined separately and the percentage increase was determined by comparing the total integrated absorption area of the dye blends with that of the individual dyes under identical measurement conditions. The areas calculated were: CL (57.535), curcumin (99.879) and BRB (103.535). When the curry leaf was co-sensitised with curcumin, the total absorption area increased to 159.52. The addition of BRB to the CL-Cur mixture resulted in an absorption area of 221.019. Another important factor of the photosensitizer dye is its absorption coefficient (α). The larger the absorption coefficient, the higher the efficiency of the DSSC. The absorption coefficient or light penetration ability of light into the dye can be determined from the Beer-Lambert law [30]:

$$I = I_0 e^{-\alpha x} \quad (2)$$

where I_0 = intensity of the incident radiation, I = intensity of the transmitted radiation; α is the absorption coefficient and x is the path length (1 cm); A is absorbance. Taking log on both sides and re-arranging, eqn. 2 yields the eqn. 3:

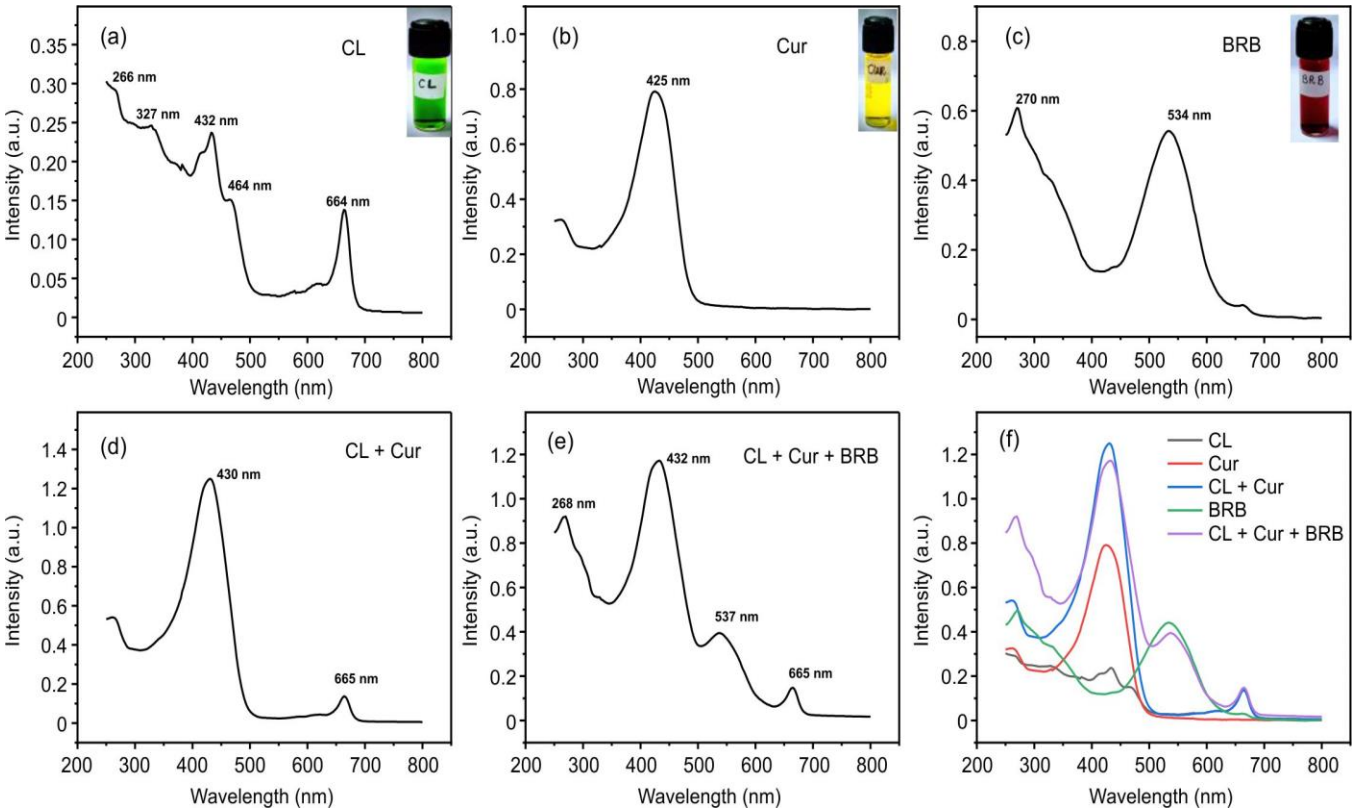

Fig. 4. Absorption spectra of (a) CL (b) Cur (c) BRB (d) CL+ Cur blend (e) CL + Cur + BRB blend (f) comparison of individual and blend dyes

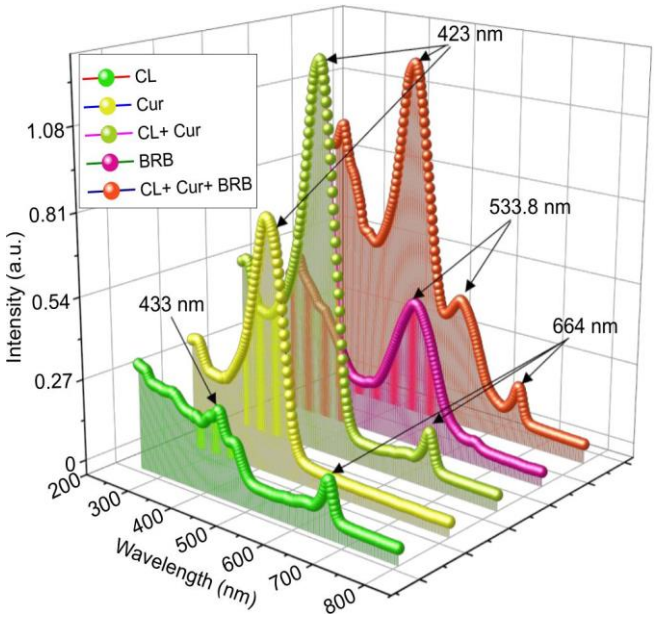

Fig. 5. UV-visible absorption spectra of single dyes (CL, Cur, BRB), bi-pigment blend (CL + Cur) and tri-pigment blend (CL + Cur + BRB)

TABLE-4
ABSORPTION COEFFICIENT AND INTEGRATED AREAS OF ABSORPTION PEAKS OF CL, CUR, BRB AND THEIR BLENDS

Dye samples	Absorption coefficient and area under each absorption curve arbitrary unit (nm)			Total area (nm)
	λ_{max} (nm)	Absorption coefficient (α)	Area (a.u., nm)	
CL	328.0	0.57	30.683	57.535
	433.0	0.55	14.783	
	463.0	0.35	5.605	
	663.0	0.32	6.464	
Cur	258.8	0.70	18.289	99.879
	329.2	0.53	3.6	
	423.0	1.80	77.99	
CL + Cur	258.8	1.25	26.369	159.52
	430.4	2.90	126.718	
	663.6	0.32	6.433	
BRB	269.8	1.14	50.195	103.535
	533.8	1.01	53.340	
CL+ Cur+ BRB	269.8	2.10	66.216	221.0197
	432.6	2.70	120.049	
	532.0	0.89	29.385	
	663.6	0.33	5.369	

$$\text{Absorption coefficient } (\alpha) = 2.303\ A\ cm^{-1} \quad (3)$$

Table-4 shows the absorption coefficients of all single and blend dyes, which were calculated from the data obtained from UV-visible absorption and eqn. 2. The absorption coefficients of the leaf extract (CL) were 0.57, 0.55, 0.35 and 0.32 for the prominent absorption regions at 328, 433, 463 and 663 nm, respectively. Upon the addition of curcumin, which has a maximum absorption at 423 nm, the absorption coefficient of CL at approximately 433 nm increased remarkably by 427% (2.9). In return, the interaction between CL and Cur increased the absorption of curcumin at 258.8 nm by 78% (1.25). Furthermore, when the BRB extract was added to the CL and Cur blend, the absorption of BRB at 269.8 nm increased by 171 % (2.1). Even though a slight decrease in the absorption of CL and Cur blend (430 nm) and BRB around 533.8 nm was observed after mixing, a prominent increase in the absorption of CL, Cur and BRB blend dye was observed, maintaining absorption at 663 nm of leaf extract unaltered in both blend dyes. The tri-pigment blend of chlorophyll, curcumin and anthocyanin enables panchromatic light harvesting [31] through complementary absorption [32] across the visible spectrum. In mixed dye systems, possible dye–dye interactions such as molecular aggregation or competitive adsorption on the TiO_2 surface may influence light harvesting behaviour [33]. However, the observed enhancement in absorption suggests that spectral complementarity between chlorophyll, curcumin and anthocyanin dominates over adverse aggregation effects. Similar behaviour has been reported in co-sensitised DSSCs, where appropriate dye combinations mitigate aggregation while improving photon absorption through complementary absorption bands [34,35]. The estimated dye concentrations obtained from the UV-visible absorption measurements indicate that comparable concentration levels were maintained during the preparation of single dyes and dye blends. This confirms that the observed enhancement in absorption for binary and tri-pigment dye blends arises primarily from spectral complementarity rather than concentration effects. The use of estimated concentrations further supports the reproducibility and consistency of the optical analysis.

The enhanced light harvesting in the tri-pigment dye blend may also be influenced by possible Förster resonance energy transfer (FRET) [36] between spectrally overlapping dyes. It may enhance overall light-harvesting efficiency and suppress charge recombination [37], thereby increasing photocurrent density and device efficiency. However, it should be noted that FRET is suggested here based on the spectral overlap considerations reported in the literature and is not experimentally verified in the present study.

Conclusion

The present study aimed to develop a promising dye that can absorb a broader range of solar radiation and enhance the efficiency of DSSCs. The selection of natural dyes based on their absorption region was the prime purpose of the investigation. Enhanced absorption of selected dyes was achieved through co-sensitisation. The blend dye comprising leaf pigment, curcumin and *Basella rubra* Berry extract has higher absorption as compared to the bi-pigment blend dye and the single pigment dye and hence the tri-pigment blend dye is expected to perform better than the single dyes in DSSC. Future work will focus on the device fabrication, photovoltaic characterisation and stability evaluation. While the present study focuses on the optical absorption enhancement, complementary analyses such as FTIR, Raman and fluorescence spectroscopy would provide deeper insight into dye-dye interactions and chemical stability of mixed dyes. These investigations will be considered in future work, along with the device fabrication and photovoltaic characterisation.

ACKNOWLEDGEMENTS

The authors acknowledge DST-FIST for providing facilities in the institution.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

DECLARATION OF AI-ASSISTED TECHNOLOGIES

During the preparation of this manuscript, the authors used an AI-assisted tool(s) to improve the language. The authors reviewed and edited the content and take full responsibility for the published work.

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