

Utilization of Mixed Naphthol Green B and Janus Green B Dyes as Photosensitier in Photogalvanic Cell for Solar Energy Conversion and Storage

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In this work, a new dye-sensitized solar cell (DSSC) was prepared using naphthol green B and Janus green B as photosensitizers and EDTA as reductant. A 210 μ A of photocurrent and 1018 mV of photo-potential were generated using these dyes. The fill factor of 0.40, a conversion efficiency was observed to be 1.0028% and the power or maximum output of cell was calculated to be 213.78 μ W. Because of the storage capacity in a cell, a DSSC can be used for a total of 180 min in dark. The properties of mixed photosensitize system EDTA-NGB-JGB were characterized and evaluated by UV-visible, fluorosencemeter, FESEM and XRD analysis. The photocurrent generation mechanism in photogalvanic cell was also discussed.

Keywords: Naphthol green B, Janus green, Conversion efficiency, Photogalvanic cell.

INTRODUCTION

An inexpensive and larger energy source is critically required for the successful growth of any developing nation. In present terms, energy is considered to be the primary element of modern era development and to cope with its need. It can be rightly said no nation could really bloom towards development without the presence of energy resources in abundance. A photoelectrochemical device used for converting light into electric energy is a simple definition of a photogalvanic cell, this also explains the battery construction which can undergo processes of discharging as well as cyclic charging [1]. When illuminated the cells are charged and the conversion of energy from light is done into chemical energy while a suitable driving reaction of redox goes on with potential gradient working against it.

Spontaneously, the reaction can also be reversed to be used in dark mode by making use of an external circuit for transferring electron. Under this method, a conversion of chemical energy is done into electric energy resulting in discharging of cell. No chemicals consumptions take place during this cycle of charge and discharge. Hence, under an ideal process, a system must work in the form of rechargeable battery with no efficiency loss as explained by Rohatgi-Mukherjee [1]. Normally, photogalvanic cells are dilute solution based dye sensitized solar power storage devices, which are quite different from other cells like galvanic or voltaic cells. Despite recent advances in photogalvanic cells leading to tremendous improvement in their electrical output [2-4], but still more efforts are required to be improve to make these cells significant pratically in daily life.

One of the biggest advantage of the photogalvanic cells over other systems is their inherent capacity of storing solar power in the form of electric energy. This can be explained as a kind of chemical usage of solar energy by means of a photogalvanic systems, employing the radiation energy from sun not being degraded into heat, although such quanta are used in both molecular and atomic systems undergoing many changes chemically. Under such chemical changes, some endergonic processes take place in the form of storage of quantum energy as labile intermediates (semi/leuco being the transition states) which are used in storing solar energy [5]. Just as photosynthesis occurs in plants, a process of photochemical conversion also works in a similar manner. On an early stages, main focus of the researchers was on enhancing the efficiency of conversion though with no energy storage this would be impossible for making it comparative to photovoltaic cells. By 1980s, few

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researchers [6-10] successfully storing the solar energy as electrical energy.

A detailed literature survey reveals that different photosensitizers (dyes) have been used in photogalvanic cells along with reductants and surfactants [11-21]. In this article, a system of EDTA-naphthol green B-Janus green B system was studied in photogalvanic cell in terms of solar power conversion and storage. A use of mixed dyes with enhanced performance of 200 min of low charging time and enhanced capacity of storage of 180 min is established.

EXPERIMENTAL

The commercial dyes, naphthol green B (NGB) and Janus green B (JGB) (Loba Chemie Pvt. Ltd., Mumbai, India) as photosensitizer, EDTA (99.8 %, Sigma-Aldrich) as reductant were used as received. The dye solutions, naphthol green B and Janus green B (M/500) and EDTA (M/100) were prepared in a double distilled water and kept in Amber coloured bottles to protect from sunlight.

Experimental set up of photogalvanic cell: A photogalvanic cell consist of thin electrochemically treated platinum foil as working electrodes, where the electrodes were SCE, a 200W Philips tungsten lamp with 10.4 mW/cm²s intensity on the cell surface was used as light source. A distance of 14 cm was fixed between the illuminated electrode and the source of light. As maximum of ultraviolet radiations were absorbed in the cell only, no UV filtration was required. However, for obstructing the infrared radiations, water filters were used.

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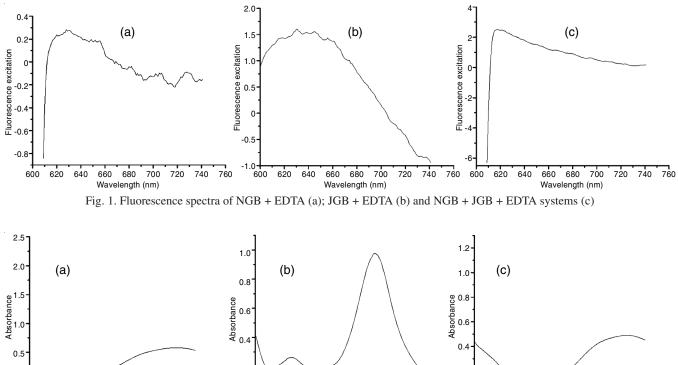
Absorption spectra of dye solutions was analyzed using UVvisible spectrophotometer (Hitachi-U3900), Spectro Fluorimeter (Agilent Carry Eclips), FTIR (Brucker Tensor 37), XRD (X-ray Rigaku Smartlab 3 kW), Field Emission Scanning Electron microscopy (Model FESEM gold coated JEOL-JEC-3000 FC).

Photochemical bleaching after illuminating, Janus green B and naphthol green B were potentiometrically studied. The generated current and potential through this system were calculated using a microammeter and a digital pH meter by making use of a key for opening one circuit and closing another. The i-V current-potential characteristic was studied by applying an external load using a variable resistor.

RESULTS AND DISCUSSION

Fluorescence analysis: A fluoroscent spectra of system EDTA-NGB, EDTA-JGB and EDTA-NGB-JGB are shown in Fig. 1. A light sensitizer is recorded at on excitation wavelength of λ_{max} 630 nm at room temperature. Among a single dye-EDTA system, a sharp change in fluorescence intensity was observed in mixed dyes with EDTA (Fig. 1c). Moreover, EDTA alone does not show any absorption in the ultraviolet range [22] and hence has almost no effect on the potential of EDTA. Thus, using a mixed organic dyes system, a red shift of absorption band and a certain degree of hypochromicity was observed.

UV-visible analysis: Absorption spectra at the optimization conditions are shown in Fig. 2. Maximum absorption in a single dye photogalvanic system (NGB-EDTA) was achieved at 703 nm. In case of JGB-EDTA system, the value was found to be



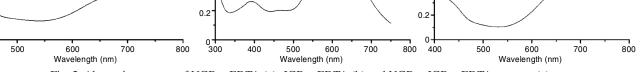


Fig. 2. Absorption spectra of NGB + EDTA (a); JGB + EDTA (b) and NGB + JGB + EDTA systems (c)

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607 nm. Therefore, a change in the wavelength absorbance was noticed in the photogalvanic system using mixed dye (NGB-JGB-EDTA) at 736 nm. An increased in the shift of the absorption spectra reflect enhancing absorption for this photosenstizer system EDTA-NGB-JGB.

FESEM analysis: In dark, photosensitizer has low conversion efficiency mainly due to inadequate absorption of photon byphotosenstizer chromophoric group in limited light exposure. Abudance of light exposure causes the photogeneration of charge carriers on the chromophore where electrons are excited [23]. The nature of dye and its photoactivity may be modulated by the photosenstizers chromophoric groups which increase the absorption on light intensity. More molecules become excited and optimized the functionalization with chromophoric photoactive groups with enhanced in solar radiation

and high electron mobility. Morphology and changed in the photoactivity of chromophoric group due to the excitation in presence of light were examined by SEM (Fig. 3). The performance of mixed dye in photogalvanic cell depends upon the molecule structure of dye, specific presence of photochromic group and the aggregation of dye molecules. The performance of NGB-JGB-EDTA system is enhanced in mixed dyes and in abudance of illumination is increased due tothe absorption of photon in NGB-JGB-EDTA system.

XRD studies: Powder XRD analyses of single dye naphthol green B, Janus green B and mixed dyes naphthol green B-Janus green B sytsems in absence and after absorption of light were analyzed (Fig. 4). The more intense XRD peaks in the mixed system confirmed that in the presence of light, absorption of photosenstizers were enhanced.

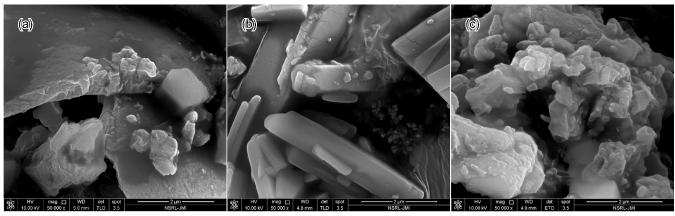


Fig. 3. SEM images of NGB + EDTA (a); JGB + EDTA (b) and NGB + JGB + EDTA systems (c)

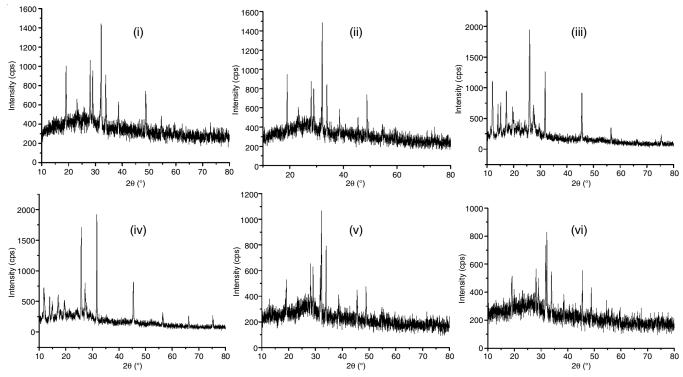


Fig. 4. XRD spectra of before absorption [NGB + EDTA (i); JGB + EDTA (ii) and NGB + JGB + EDTA (iii)]; and after absorption of light [(NGB + EDTA (iv); JGB + EDTA (v) and NGB + JGB + EDTA systems (vi)]

Effect of pH: Consequence of a pH value can also be observed on the back reaction, hence a suitable value of pH could also help in increasing the output of the electrical power. Generally, it was noticed that an output of photogalvanic cell regeneration rises when alkali concentration enhanced and then goes on decreasing, once reached a maximum value. An optimum condition of pH value is dependent in the reductant pKa value. It was elaborated under a mere fact that reductant is found only in an anionic form and consider as a better form of donor [24].

The cell outputs as an electric signal and can affect in terms of pH variation. With a further increase in pH, an output of electrical signal also increases (Table-1). Maximum value of photo-current and photo-potential were achieved at pH 12.0, although an additional increase in pH value leads to a decreasing electrical output (Fig. 5). Hence, the photogalvanic cell with EDTA-naphthol green B and Janus green B are highly sensitive to the solution. Another observation was made that an optimum condition of pH 12.8 is in relation to the reductant pKa where desired value of pH in more than pKa (pH > pKa).

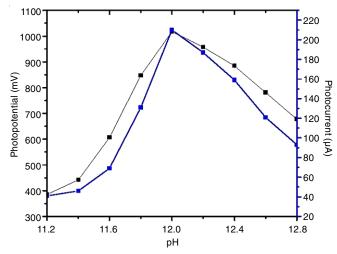


Fig. 5. Variation of the photocurrent and photopotential with pH solutions for NGB + JGB + EDTA system

Effect of EDTA concentration: In this system, EDTA was used as an irreversible reductant which could be used in the form of electrolyte. A cell produces an electric output can have

impact due to EDTA concentration variations. The results are represented in Fig. 6 and Table-2. The reducing agents with low concentrations decreases electrical power, due to the lesser number of molecules present in the reducing agent. The reducing agent with high concentrations also reduced electrical power, due to the increases number of the reducing agent molecules affects the molecules of dye to reaching at the electrodes and observation was made that an optimum condition of maximum electrical output recorded for system at 9.3×10^{-3} M.

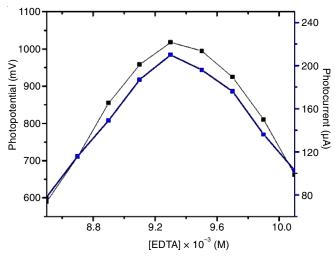


Fig. 6. Variation of EDTA concentration with photocurrent and photopotential for NGB + JGB + EDTA system

Effect of mixed dyes concentration: Photosensitizer when used in lower concentrations, results in lower concentrations of photocurrent and photo-potential due to a less molecules of photosensitizer present for consecutive excitation or donation of electrons to an platinum electrode. In a similar manner, when used higher concentrations, maximum light is absorbed by the dye molecules present in a path as photogalvanic cells, which is controlled by diffusion [25]. The higher concentration might results in higher recombination occurring in dye and the injected electrons. Hence, an amplified photocurrent and photo-potential with increasing photosensitizer concentration attain to a maximum value and then decreased.

The photocurrent and photo-potential dependence on the dyes concentration was conducted and the outcomes are shown

TABLE-1 EFFECT OF VARIATION OF pH									
$[NGB] = 4.22 \times 10^{-5} \text{ M}; [EDTA] = 9.3 \times 10^{-3} \text{ M}; [JGB] = 1.53 \times 10^{-5} \text{ M}; \text{ Light intensity} = 10.4 \text{ mW cm}^2; \text{ Temp} = 303 \text{ K}$									
pH 11.2 11.4 11.6 11.8 12.0 12.2 12.4 12.6								12.8	
Photoptential (mV)	386	442	607	848	1018	958	885	781	678
Photocurrent (µA)	41	46	69	131	210	187	159	121	93
Power (µW)	15.82	20.33	41.88	111.08	213.78	179.14	140.71	94.50	63.05

TABLE-2 EFFECT OF VARIATION OF EDTA CONCENTRATION									
$[NGB] = 4.22 \times 10^{-5} \text{ M}; [JGB] = 1.53 \times 10^{-5} \text{ M}; \text{ pH} = 12.0; \text{ Light intensity} = 10.4 \text{ mW cm}^2; \text{ Temp.} = 303 \text{ K}$									
[EDTA] × 10 ⁻³ M	8.5	8.7	8.9	9.1	9.3	9.5	9.7	9.9	10.1
Photoptential (mV)	590	711	855	958	1018	995	925	811	663
Photocurrent (µA)	78	116	149	187	210	196	176	136	103
Power (µW)	46.02	82.47	127.39	179.14	213.78	195.02	162.8	110.29	68.28

in Table-3. A dyes in lower concentration ([JGB] < 1.53×10^{-5} M, [NGB] < 4.22×10^{-5} M) results in low photocurrent and photo-potential due to the lower number of molecules in dye present for consecutive donation and excitation of electrons to platinum electrode (Fig. 7a). Dyes in higher concentration ([JGB] < 1.53×10^{-5} M, [NGB] < 4.22×10^{-5} M) results in low electric output, because of the light intensity which reach the molecules of dyes near an electrode lowers because maximum part of lights is absorbed by the molecules of dyes (Fig. 7b).

Electroactive species: Different methods could be used for generating photocurrent in the cells. The combinations possible in the photogalvanic cell electroactive species are stated below:

In the illuminated chamber	In the dark chamber				
NGB + JGB	Oxidized form of reductant (R ⁺)				
Leuco or Semi-NGB ⁻ + JGB ⁻	Oxidized form of reductant (R ⁺)				
Leuco or Semi-NGB ⁻ + JGB ⁻	NGB + JGB				

Oxidized reductants are formed only in an illuminated chamber, where in case of dark chambers these are considered as electroactive species, these must diffuse into a dark chamber from an illuminated chamber for accepting the electrons from electrodes. As a result, the maximum photocurrent (i_{max}) as well as the photocurrent generation must decrease with increasing diffusion length, however its was not confirmed experimentally. At equilibrium the photocurrent value (i_{eq}) was noticed to be independent with respect to changing length of diffusion

A 01.0028% of conversion efficiency were achieved by determining it with a 200W lamp working as thesource of light as this has around 15-times lower light intensity as that of the direct light from sun. Hence, an efficiency of conversion is observed 15.042% experimentally. The photogalvanic cells shows lower levels of conversion efficiency when compared to a 18% of the theoretical value. This is because of the dye molecules aggregation around an electrode, dyes with lower stability, and the transfer back-electron, *etc.* It is advantageous to use the electrochemical device for investigating in terms of 180 min of storage or charging time or a better value of electrical output.

Mechanism: The energy storage mechanism is explained by using the life time of leuco/semi of dye as well as the transfer rate of electrons between the reducing agent and the dye. The photogalvanic cell show better capacity of storage in case of more stable excited molecules of dye because of their bulkiness or the number of excited electrons delocalized on it or for a reason like the diffusion resulting in recombination, the reduced dye concentration and lastly because of reduced capacity of

TABLE-3 EFFECT OF NAPHTHOL GREEN B (NGB) AND JANUS GREEN B (JGB) CONCENTRATION									
[ED	$[EDTA] = 9.3 \times 10^{-3} \text{ M}; [JGB] = 1.53 \times 10^{-5} \text{ M}; \text{ Light intensity} = 10.4 \text{ mW cm}^2; \text{ pH} = 12.0; \text{ Temp.} = 303 \text{ K}$								
$[NGB] \times 10^{-5} M$ 3.42 3.62 3.82 4.02 4.22 4.42 4.62 4.82 5.02									
Photoptential (mV)	627	737	855	981	1018	958	812	627	590
Photocurrent (µA)	93	121	146	187	210	168	122	93	84
Power (µW)	58.31	89.17	124.83	183.44	213.78	160.94	98.25	58.31	49.56
[NG	$[NGB] = 4.22 \times 10^{-5} \text{ M}; [EDTA] = 9.3 \times 10^{-3} \text{ M}; \text{ pH} = 12.0; \text{ Light intensity} = 10.4 \text{ mW cm}^2; \text{ Temp.} = 303 \text{ K}$								
$[JGB] \times 10^{-5} M$	0.73	0.93	1.13	1.33	1.53	1.73	1.93	2.13	2.33
Photoptential (mV)	516	590	774	958	1018	922	796	737	700
Photocurrent (µA)	74	93	121	179	210	159	131	116	103
Power (µW)	38.18	54.87	93.65	170.52	213.78	146.59	104.27	85.49	72.1

in their path.

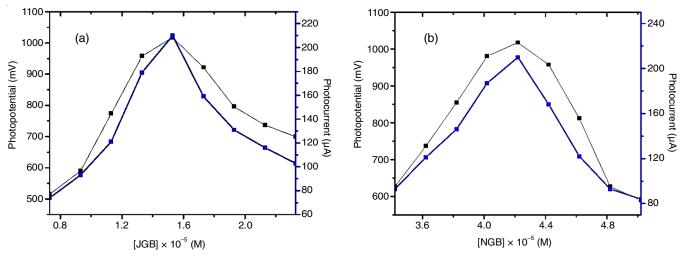


Fig. 7. Variation of JGB (a) and NGB (b) concentration with photocurrent and photopotential for NGB + JGB + EDTA system

storage in the photogalvanic cell. As per the investigation presented above the photocurrent generation mechanism in photogalvanic cells are proposed as given below:

Illuminated chamber: The molecules of dye get excited on irradiation

$$NGH + JGB \xrightarrow{h\nu} NGB^* + JGB^*$$
(3)

The excited molecules of dye accept the single electron through the reductant and is converted into the form of leuco and semi of dyes and oxidized form of reductant:

$$NGB^* + JGB^* + 2R \longrightarrow NGB^- + JGB^- + 2R^+ \qquad (4)$$

Working electrode: Leuco or semi forms loses an electron and converts it into the molecules of the original dye:

$$NGB^- + JGB^- \longrightarrow NGB + JGB + 2e^-$$
(5)

Dark chamber

Counter electrode: The molecules of the dye accept an electron from its electrode and converts in leuco or semi forms:

$$NGB + JGB + 2e^{-} \longrightarrow NGB^{-} + JGB^{-}$$
(6)

Conclusively, dye semi/leuco form and oxidized reductant forms for generating the reductant molecules and original dyes:

 $NGB^{-} + JGB^{-} + 2R^{+} \longrightarrow NGB + JGB + 2R$ (7)

Herein, NGB + JGB , PB* + NGB* ,PB⁻ + NGB⁻, R and R⁺ are their excited forms leuco or semi-leuco forms, reductant and R⁺ its oxidized forms, respectively

Effect of diffusion length: The effects of diffusion length variations on the parameters of current viz., i_{eq} , i_{max} and the initial current generation rate for cells were studied by using different dimensions of H-cells. Results are given in Table-4. It was noticed that photocurrent (i_{max}) increased sharply for initial few illumination minutes and then the photocurrent decreases gradually to a more stable value (i_{eq}). On illumination, a spontaneous rise in photocurrent results in an initial rapid reaction with additional step of determining slow rate on last step.

TABLE-4 EFFECT OF DIFFUSION LENGTH									
$[NGB] = 4.22 \times 10^{-5} \text{ M}; [EDTA] = 9.3 \times 10^{-3} \text{ M}; [JGB] = 1.53 \times 10^{-5} \text{ M}; \text{ Light intensity} = 10.4 \text{ mW cm}^2; \text{ pH} = 12.0; \text{ Temp.} = 303 \text{ K}$									
Diffusion	Diffusion Maximum Equilibrium Rate of initial								
length (DL,	-								
mm)	mm) $(i_{max}, \mu A)$ $(i_{eq}, \mu A)$ current $(\mu A \min^{-1})$								
35.0	229	202	14.8						
40.0	238	208	15.9						
45.0	249	210	17.0						
50.0	257	208	18.1						
55.0	265	206	19.2						

Effect of electrode area: The current parameters of a cell and its impact on the electrode area was also studied. The effects of variation of platinum electrode area on the cell were studied by constructing four photogalvanic cells while keeping all the other factors optimum. It was found that that maximum photocurrent (i_{max}) increases with an increase in electrode area, where as photocurrent (i_{eq}) at equilibrium was observed to be independent of change in the electrode area (Table-5).

Current potential (i-V) characteristics of cell: The i_{sc} short circuit current was found to be 210 μ A, while the V_{oc} open circuit voltage was 1248 mV for the photogalvanic cells. The values were calculated using a digital pH meter with an open circuit and a microammeter with closed circuit, respectively. The potential as well as current values lie in the two recorded extreme ranges using a 500 K carbon pot log resistor in the microammeter circuit by applying an external load.

Power point (pp) is the determined point on the i-V curve, where the product of current (i_{pp}) and potential (V_{pp}) were maximum. By using i-V curve, an estimated fill factor was found out to be 0.40 and a 1.0028 % of conversion efficiency. Here, fill factor (FF) of a cell is another crucial parameter used to measure the performance, which represents the value of how much short circuit current and open circuit voltages are obtained on the maximum value of power.

The calculation of conversion efficiency for the photogalvanic cells was performed using power point output and an incident radiation power. At an optimum condition, a system was under the exposure of sunlight. It can be said that a deviation of i-V curve from a rectangular curve shape as depicted in Fig. 8.

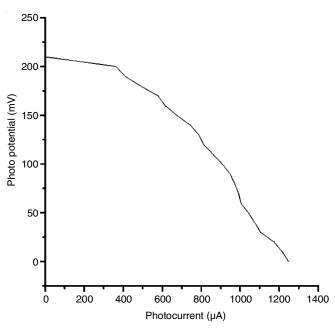


Fig. 8. Current potential (i-V) curve of the photogalvanic cell using NGB + JGB + EDTA system

TABLE-5 EFFECT OF ELECTRODE AREA								
$[NGB] = 4.22 \times 10^{-5} \text{ M}; [EDTA] = 9.3 \times 10^{-3} \text{ M}; [JGB] = 1.53 \times 10^{-5} \text{ M}; \text{ Light intensity} = 10.4 \text{ mW cm}^2; \text{ pH} = 12.0; \text{ Temp.} = 303 \text{ K}$								
Electrode area (cm ²)	0.36	0.64	1.00	1.44	1.69			
Maximum photocurrent (i _{max} , µA)	226	230	234	239	241			
Equilibrium photocurrent (i_{eq} , μA)	206	208	210	211	208			

Performance of the cell: The photogalvanic cell has an additional advantage of being used in dark. The capacity of storage of photogalvanic cells was checked by using an external load required for power point current after terminating the illumination when the potential was reached to a constant value. The capacity of storage was determined by $t_{1/2}$, where $t_{1/2}$ is a time required in decreasing power output to half of the power point in dark. In present work, a system of natural dyes (naphthol green B and Janus green B) mixed with EDTA in the photogalvanic cell were investigated. An observation is made that a cell could be be active for 180 min.

Conclusion

In this work, the results reveals that the electrical output increased by 1.5-2.0 times by using a combination of two different mixed photosensitizer dyes (naphthol green B and Janus green B). System of mixed dye when applied in this research experiment, shows enhanced electrical output, with a better efficiency of energy conversion and better capacity of storage. This kind of combination results in a promising alternative for the existing system using dyes in the development of photogalvanic cells.

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CONFLICT OF INTEREST

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

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