Brazilian Journal of Physics (2021) 51:1459–1465
https://doi.org/10.1007/s13538-021-00970-z

CONDENSED MATTER

Dye-Sensitized Solar Cells Using a Cocktail of Synthetic (Eosin Y) and Natural (Beetroot, Pomegranate, and Kumkum) Dyes

Ruba N[1] · Sowmya S[1] · Pooja Prakash[1] · Janarthanan B[1] · Nagamani Prabu A[1]

Received: 3 February 2021 / Accepted: 28 July 2021 / Published online: 6 August 2021

Abstract
For the purpose of the study, dye extracts were prepared from natural sources like beetroot, pomegranate, kumkum, and separately mixed with Eosin Y dye. TiO_2 nanoparticles were coated over the fluorine-doped tinoxide (FTO) by doctor blade technique, and a counter cathode was prepared by coating the graphene over FTO film. A sandwich-type solar cell was made and the photovoltaic performance measured using Keithley Electrometer 6517B with a Xenon lamp of 100 mW/cm^2 as solar simulator. Among the cocktail of dyes, solar cells with beetroot and Eosin Y dye exhibited the highest efficiency of 0.3%. The conversion efficiency of solar cell with pomegranate and Eosin Y was 0.1%, while that of the cells with kumkum and Eosin Y was 0.2%.

Keywords DSSC · Eosin Y · Cocktail · Natural dyes · Power conversion efficiency

1 Introduction

As one of the nonconventional energy resources, solar energy has the inimitable advantages of other energy sources that are clean and inexhaustible. Solar cells are the most efficient means of utilizing the solar energy, and currently, silicon solar cells are commercialized for the generation of power. For some reason, the availability of silicon solar cells is limited, and therefore, researchers are constrained to develop an alternative to it [1]. The dye-sensitized solar cell (DSSC) has received much attention because of its stability and efficiency. The maximum efficiency obtained for the DSSC is 14% [2]. The main advantages of DSSC are its low cost and productivity under laboratory conditions.

The structure of the DSSC consists of photoanode (transparent conductive glass sheet, semiconductor oxide, and sensitizer), counter electrode (typically Pt-coated transparent conductive glass sheet), and electrolyte. DSSCs are fabricated with two sheets of transparent conductive materials on which the semiconductor oxide and catalyst are deposited, and these act as electrodes [3]. The transparent conductive substrate should possess more than 80% of transparency to pass the optimum level of light to the active area of the solar cell and high conductivity for an effective charge transfer. Photoanodes are synthesized by depositing a layer of semiconductor materials such as TiO_2, SnO_2, ZnO, and Nb_2O_5 on glass plate coated with fluorine-doped tinoxide (FTO) or indium tinoxide (ITO). Dye is the key component for absorption of the incident light so that the absorption spectra of the sensitizer cover the UV–visible and infrared region. According to the structure, dyes are divided in two types, namely, organic and inorganic. Inorganic dyes include ruthenium and osmium complexes, inorganic quantum dots, phthalocyanine and metal porphyrin, and organic dye cover natural and synthetic sensitizers. Counter cathodes are usually prepared by platinum or carbon materials. Typically, platinum is used as counter cathode due to its higher efficiency. But cost wise Pt is not feasible for the commercialisation of DSSC. To replace the Pt, many new types of counter electrodes have been developed [4, 5]. To regenerate the oxidized dye, redox couple is used in DSSC. Since the beginning of DSSC development (I^-/I_3^-), redox couple has been preferred due its rapid dye generation, good solubility, suitable redox potential, and non-absorption of too much sunlight. At first, light is absorbed by a dye molecule, and electrons get stimulated from the ground state to the excited state of the sensitizer. Now the electrons are injected to the conduction band (CB) of semiconductor, and as a result, the dye molecules get

✉ Ruba N
rubanatarajan5@gmail.com

[1] Department of Physics, Karpagam Academy of Higher Education, Eachanari Post, Pollachi Main Road, Tamilnadu, India

oxidized. These electrons reach up to the load and counter electrode. The counter cathode acts as a catalyst, and reduction of electrolytes takes place. The redox mediator completes the circuit by regenerating the oxidized dye.

Distinct types of sensitizers are used in the DSSC. The metal free organic sensitizers have many advantages over the other sensitizers such as inexpensiveness, broad resources, and eco-friendliness. Eight different organic dyes have been prepared to fabricate a DSSC, and the highest efficiency is obtained by the Eosin Y sensitizer [6, 7]. The betanin pigment is extracted from beetroot using medium pressure liquid chromatography, and 2.7% of efficiency is reported [8]. The PCE of 2% is achieved for the solar cell with pomegranate dye extract [9]. To fabricate a solar cell, the cocktail dye of Eosin Y and hibiscus sabdariffa is prepared and 2.02% is achieved for the dye [10]. A mixture of Eosin Y and rose Bengal has been used as sensitizer in dye sensitized solar cell and power-conversion efficiency of 1.74% is reported [11].

In this work, the efficient organic dyes were chosen for DSSC and a cocktail of the organic dyes were synthesized, and the cocktail contained synthetic and natural dyes. The chosen dyes were Eosin Y, beetroot, pomegranate, and kumkum. For the first time to our knowledge, a combination of these dyes was used in the fabrication of DSSC.

2 Experimental Procedure

2.1 Preparation of Sensitizers

Eosin Y dye solution was prepared by stirring 10 mg of dye powder in 25 ml of ethanol for 15 min and kept at room temperature for 24 h. For betalain dye preparation, 15 g of fresh beetroot was crushed in a mortar and 20 ml of ethanol was added to the crushed sample. Similarly, anthocyanin pigment was extracted from pomegranate fruit. For kumkum dye preparation, 1 g of calcium carbonate was added to 15 g of turmeric, and ethanol was added to the mixture. After a day, the solutions were filtered out to remove solid residues. The cocktail dyes were formed by adding Eosin Y into natural (beetroot, pomegranate, and kumkum) dyes separately.

2.2 Preparation of Photoanodes

In this work, fluorine-doped tinoxide was used as substrate for the electrodes. In order to form a semiconductor oxide-paste, nitric acid and acetic acid were added to the titanium dioxide nano powder, and the resulting paste was deposited on the FTO substrates by doctor blade method. TiO_2 films were annealed at 450 °C for 30 min and sensitized by immersing the substrates in the dye solution for 24 h.

2.3 Device Fabrication

The immersed TiO_2 films in the dye solution were removed and rinsed with distilled water. The counter cathode was prepared by coating the graphene over the FTO substrate. For electrolyte preparation, the mixture of lithium iodide and iodine was dissolved in acetonitrile solvent. The TiO_2 films and counter cathodes were assembled into sandwich type solar cells, and a drop of electrolyte solution was introduced between the electrodes. Prepared dyes and fabricated cells are shown in Fig. 1.

3 Result and Discussion

3.1 Absorption Spectra

The absorption spectra of the cocktail dyes were taken using ultraviolet visible spectrometer in the wavelength range from 200 to 850 nm as shown in Fig. 2. The absorption peak of Eosin Y was found to be 520 nm, which matches with the previous record reported by Amal M. Al-kahlout et al. [6]. Betalain extract from beetroot shows an absorption peaks at 477 nm and 537 nm which is similar to the value reported by S. Sathyajothi et al. [12]. The cocktail of Eosin Y and beetroot dye has two peaks centred at 483 nm and 520 nm. The first peak represents the absorption of beetroot, and the second indicates the absorption range of the Eosin Y dye as a single dye. Anthocyanin extract from pomegranate displayed two peaks at 443 nm and 510 nm that matches with the work of William Ghann et al. [9]. The mixture of pomegranate and Eosin Y has absorption peaks centred at 500 nm and 525 m. The peak at 500 nm indicates the presence of pomegranate, and the peak at 525 nm shows the absorption of Eosin Y. In the case of cocktail dye of kumkum and Eosin Y, the first peak is observed at 415 nm and the second at 525 nm. The absorption peak of kumkum dye is identified at 413 nm. All the cocktail dye solutions showed a wider range of absorption in the visible region. It can be seen that cocktail dye possesses the characteristics of both the constituents of synthetic and natural dyes in the visible region.

S. no	Dye extracts	Absorption peaks (nm)	Bandgap (eV)
1	Beetroot	477	2.6
		537	2.3
2	Eosin y + beetroot	483	2.6
		520	2.4
3	Pomegranate	443	2.8
		510	2.4

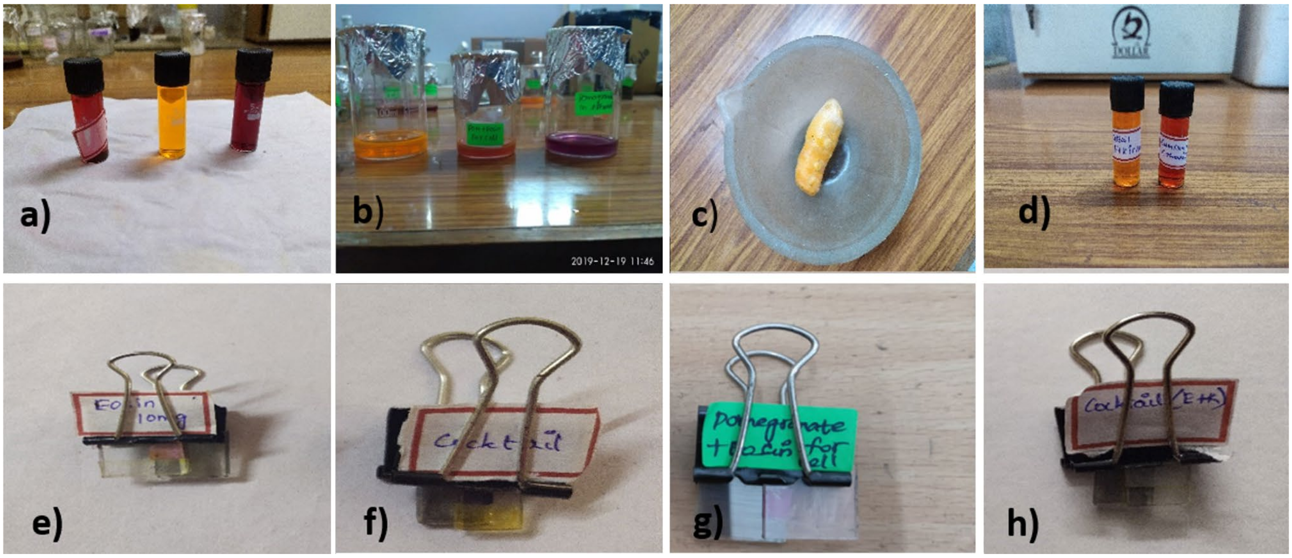

Fig. 1 Prepared dyes and fabricated solar cell: **a** cocktail of Eosin Y and beetroot, **b** cocktail of Eosin Y and pomegranate, **c** fresh turmeric, **d** cocktail of kumkum and Eosin Y, **e** DSSC with Eosin Y dye, **f** DSSC with Eosin Y and beetroot, **g** DSSC sensitized Eosin Y with pomegranate, and **h** DSSC sensitized kumkum and Eosin Y

S. no	Dye extracts	Absorption peaks (nm)	Bandgap (eV)
4	Eosin y + pomegrante	500	2.5
		520	2.4
5	Kumkum	413	3
6	Eosin Y + kumkum	415	3
		525	2.4
7	Eosin Y	520	2.4

3.2 FTIR Spectra

To fabricate an efficient solar cell, a dye should be absorbed on the surface of a semiconductor oxide so that the dye possesses a functional group like CO, OH, and COOH. Figure 3 shows the FTIR spectra of the co-sensitized dye and single dye to indicate the functional groups present in the dye solution. A wide band is observed between 3300 to 3800 cm^{-1} for all the dyes, which proves the presence of OH group in dye solutions. The peaks observed at around 2970 cm^{-1} and 2880.22 cm^{-1} are due to the OH stretching vibrations. In the spectrum, the broad band at around 1045 cm^{-1} was observed due to CO stretching vibrations and the stretching vibrations appeared at around 1650 cm^{-1}, which also indicated the CO group. The presence of CO and OH groups in the dye solution was confirmed by the FTIR spectrum.

3.3 J-V characteristics of DSSC

The cells were fabricated by the combination of Eosin Y with different natural sensitizers, and the JV curve is shown in Fig. 4. The performance of the cell was evaluated by open circuit voltage (V_{oc}), short circuit current density (J_{sc}), fill factor (FF), and power conversion efficiency (η) and listed in the Table 1. V_{oc}, J_{sc}, FF, and η obtained in our DSSC using Eosin Y are 0.5 V, 2.02 mA/cm^2, 0.36, and 0.4% respectively, and the PCE of the Eosin Y dye was higher than the previous records [6, 13, 14]. The PCE of the solar cell prepared from the cocktail dye of beetroot and Eosin Y is 0.3%, with V_{oc} of 0.3 V, J_{sc} of 2.84 mA/cm^2, and FF of 0.35. The conversion efficiency of 0.1% was recorded for the mixture of pomegranate and Eosin Y with V_{oc} of 0.13 V, J_{sc} of 2.3 mA/cm^2, and FF of 0.25. In the case of kumkum and Eosin Y cocktail, the cell exhibited 0.2% of PCE with V_{oc} of 0.2 V, J_{sc} of 2.86 mA/cm^2, and FF of 0.3. From the results, it can be observed that the solar cell with the cocktail dye of beetroot and Eosin Y exhibited the highest efficiency compared to other cocktail dyes. But the efficiency of Eosin Y is higher than the cocktail dyes. However, the efficiency value of cocktail of beetroot and Eosin y is still closer to the efficiency of Eosin y. Hence, the combination of Eosin Y with natural dyes is the promising work for DSSC.

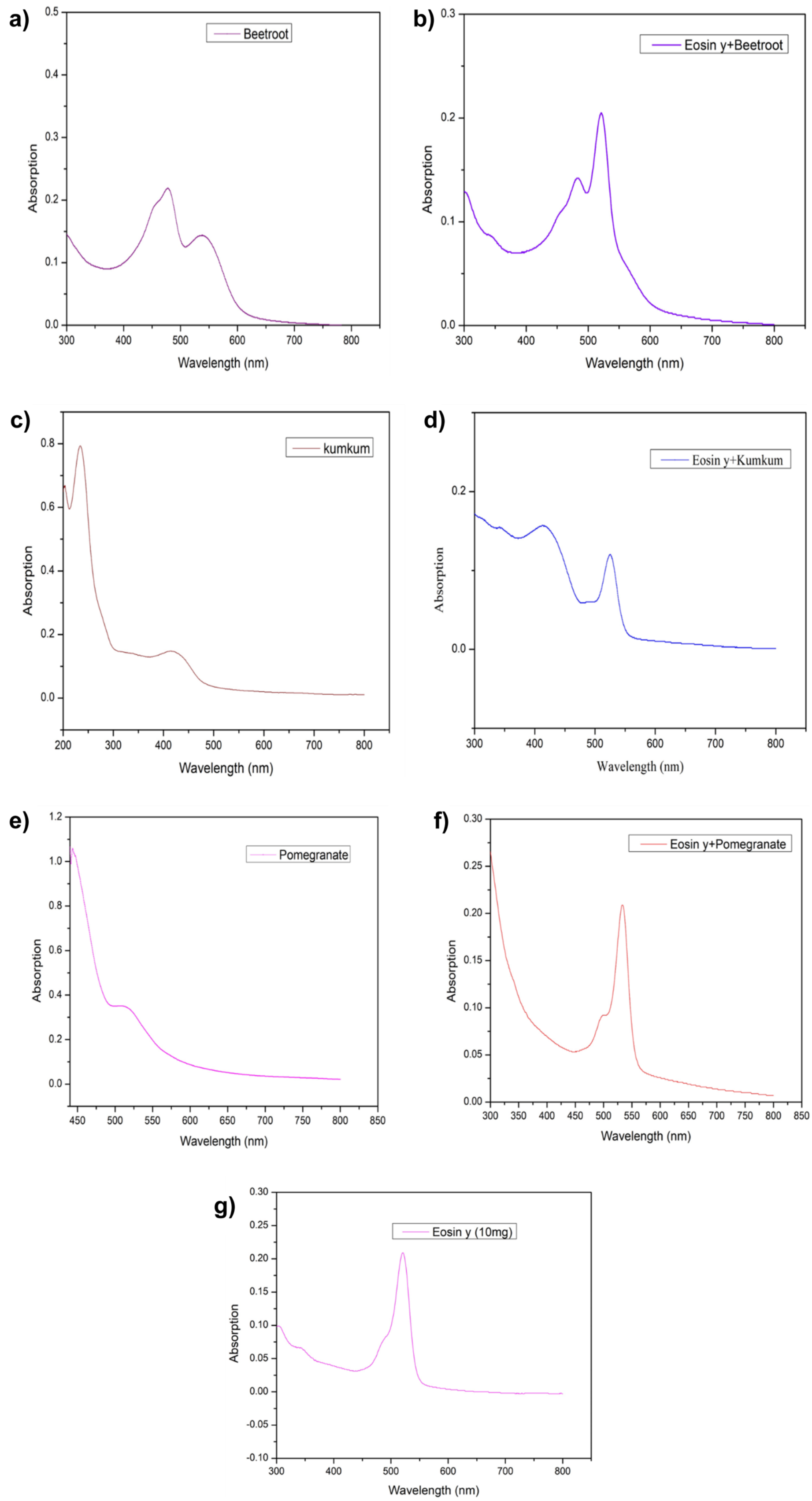

Fig. 2 Absorption spectra of single and cocktail dye solutions **a** beetroot, **b** Eosin y + beetroot, **c** kumkum, **d** Eosin Y + kumkum, **e** pomegranate, **f** Eosin Y + pomegranate, and **g** Eosin Y

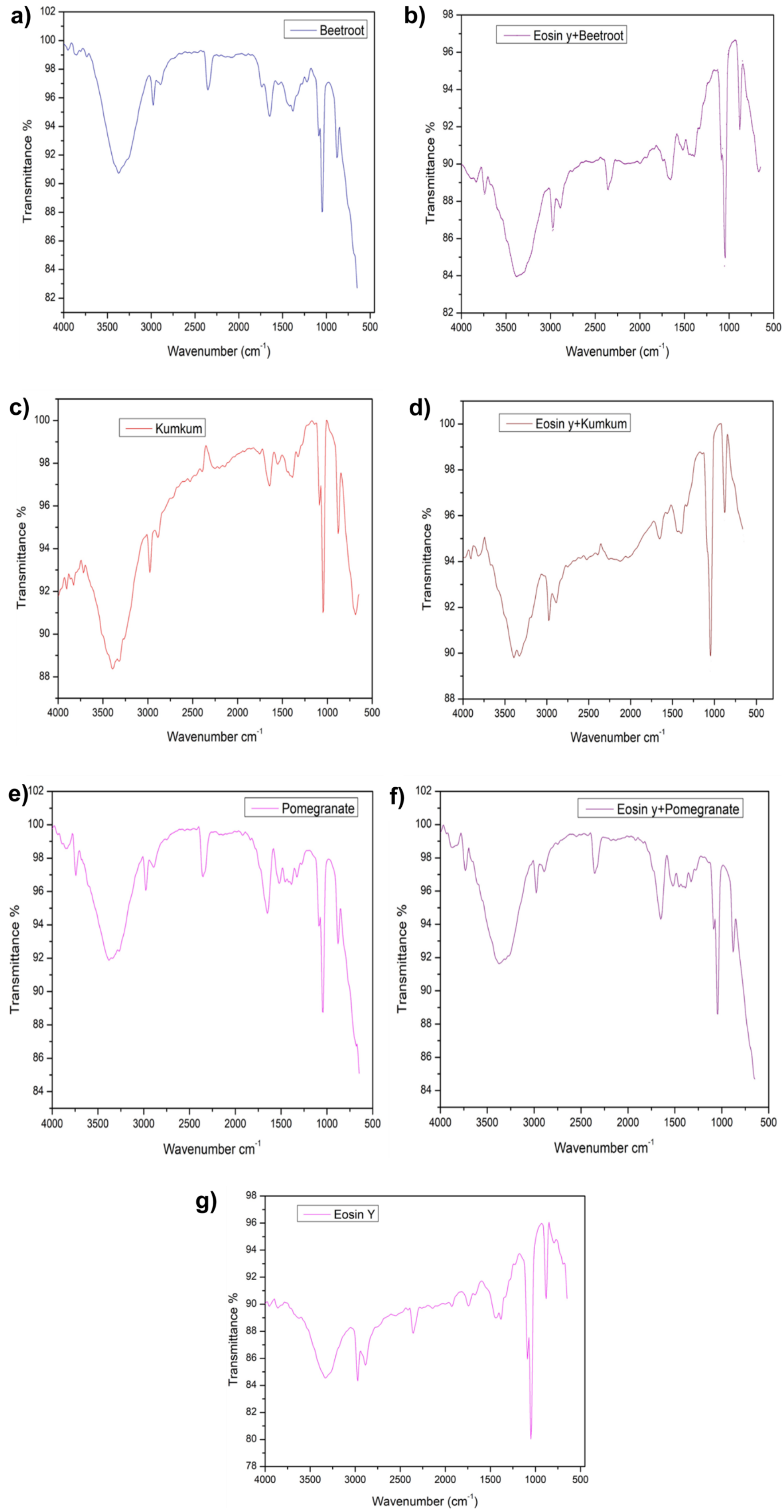

Fig. 3 FTIR spectrum of single and cocktail dyes: **a** beetroot, **b** Eosin Y + beetroot, **c** kumkum, **d** Eosin Y + kumkum, **e** pomegranate, **f** Eosin Y + pomegranate, and **g** Eosin Y

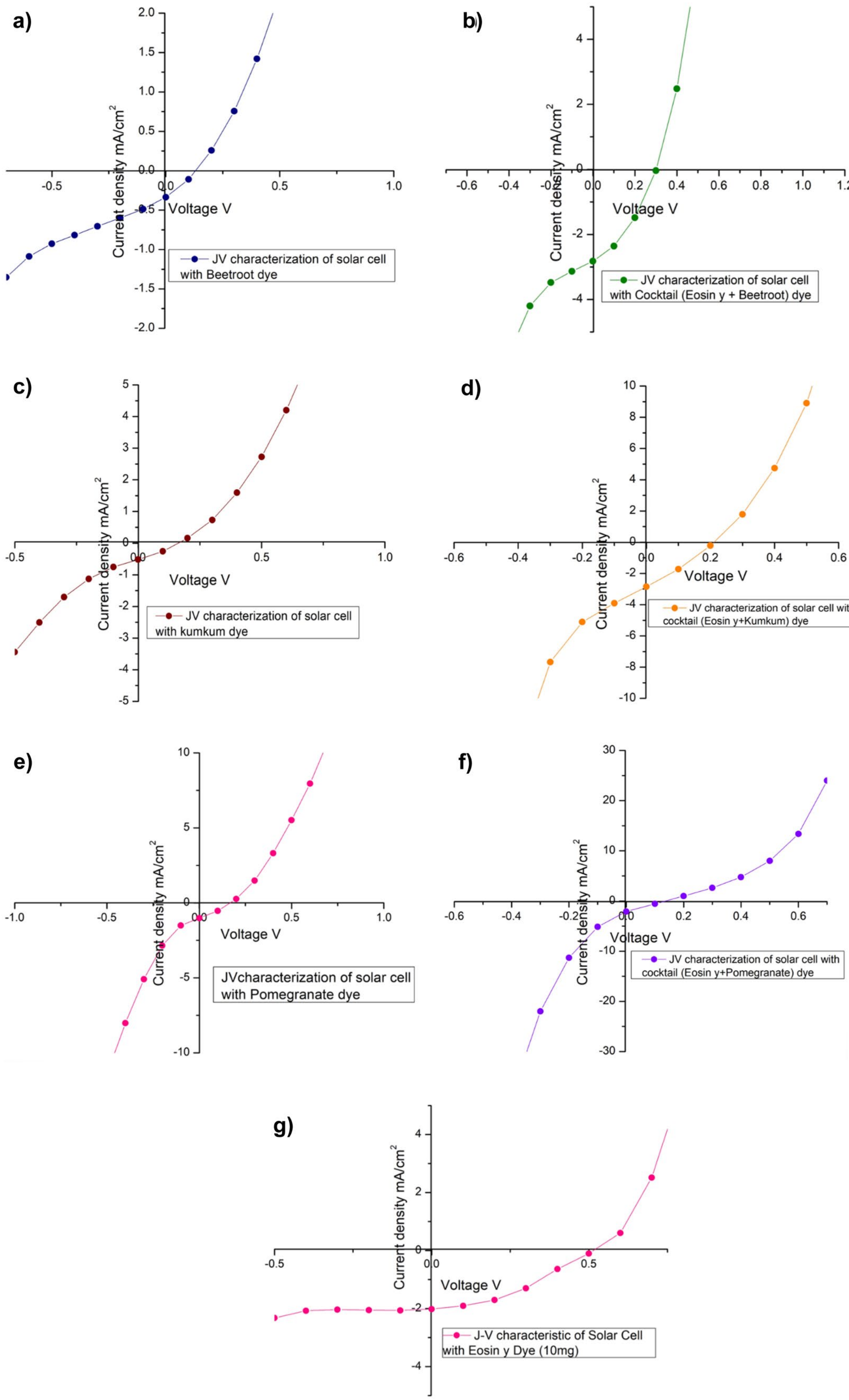

Fig. 4 J-V characteristics of DSSC with **a** beetroot, **b** Eosin Y + beetroot, **c** kumkum, **d** Eosin Y + kumkum, **e** pomegranate, **f** Eosin Y + pomegranate, and **g** Eosin Y

Table 1 The performance of the cell evaluated by open circuit voltage (V_{oc}), short circuit current density (J_{sc}), fill factor (FF), and power conversion efficiency

Sensitizer	Voltage (V_{oc}), V	Current density (J_{sc}), mA/cm^2	Fill factor (FF)	Efficiency (%)
Eosin Y	0.5	2.02	0.36	0.4
Beetroot	0.12	0.33	0.32	0.01
Pomegranate	0.17	1	0.25	0.05
Kumkum	0.15	0.04	0.5	0.03
Eosin Y + beetroot	0.3	2.84	0.35	0.3
Eosin Y + kumkum	0.2	2.86	0.3	0.2
Eosin Y + pomegranate	0.13	2.3	0.25	0.1

4 Conclusion

The solar cells were fabricated using different combinations of Eosin Y with natural dyes such as beetroot, pomegranate, and kumkum separately. UV–visible spectra provide information about the range of absorption so that the UV–visible spectra were taken for all the prepared dye solutions, and the functional groups of the dyes were analysed by FTIR spectrum. The UV–visible spectra of the dyes confirmed the broad range of absorption in the visible region, and the FTIR spectrum proves the presence of the CO and OH group in dye solutions. Compared to all the cocktail dyes, the Eosin Y with beetroot-dye-sensitized-DSSC showed better efficiency of 0.3%, and the efficiency of Eosin with pomegranate was 0.1% and the efficiency of Eosin with kumkum dye 0.2%. The future work will be proceeded with the cocktail of Eosin Y and efficient natural dyes for DSSC.

Declarations

Conflict of interest The authors declare no competing interests.

References

1. ChuanfaZhang DailinLi, NingWang, YangYu, DanYang, HuafengZhu, Guilei Li, A novel way for improving the overall photoelectric conversion efficiency of dye sensitized TiO2 solar cell. Procedia Computer Science **131**, 1336–1345 (2018)
2. Siva Sankar Nemala, Purnendu Kartikay, Rahul Kumar Agrawal, Parag Bhargava, Sudhanshu Mallick, Sivasambu Bohma, Few layers graphene based conductive composite inks for Pt free stainless steel counter electrodes for DSSC. Sol. Energy **169**, 67–74 (2018)
3. U. Mehmood, S. Rahman, K. Harrabi, I.A. Hussein, B.V.S. Reddy, Recent advances in dye sensitized solar cells, Advances in Materials Science and Engineering. Article ID **974782**, 1–12 (2018)
4. A.Sarkar Amit, K. Chakraborty, S. Bera, NiS/rGO nanohybrid: an excellent counter electrode for dye sensitized solar cell, Solar Energy Materials and Solar Cells **182**. 314–320, (2018)
5. C. Ravi Dhas, A. Jennifer Christy, R. Venkatesh , Esther Santhoshi Monica S, Subhendu K. Panda, B. Subramanian , K. Ravichandran , P. Sudhagar , A. Moses Ezhil Raj, Low-cost and eco-friendly nebulizer spray coated $CuInAlS_2$ counter electrode for dye-sensitized solar cells, Physica B: Condensed Matter **537,** 23–32 (2018)
6. M. Amal Al-Kahlout, S. Hatem El-Ghamri, Naji Al Dahoudi, M. Taher El-Agez, A. Sofyan Taya, S. Monzir, A Abdel-Latif, A comparative study: synthetic dyes as photosensitizers for dye-sensitized solar cells, Turkish Journal of Physics **39,** 272–279 (2015)
7. M. Taher El-Agez, A. Sofyan Taya, S. Kamal Elrefi, S. Monzir, Abdel-Latif, Dye-sensitized solar cells using some organic dyes as photosensitizers. Optica Applicata 2 (2014). https://doi.org/10.5277/oa140215
8. C. Sandquist, J.L. Mc-Hale, Improved efficiency of betanin-based dye-sensitized solar cells. Journal of Photochemistry Photobiology A: Chemistry **221**, 90–97 (2011)
9. W. Ghann, H. Kang, T. Sheikh, S. Yadav, T. Chavez-Gil, F. Nesbitt, J. Uddin, Fabrication, optimization and characterization of natural dye sensitized solar cell, Scientific Results (2017)
10. G. Richhariya, A. Kumar, Fabrication and characterization of mixed dye: natural and synthetic organic dye. Opt. Mater. **79**, 296–301 (2018)
11. S. Shubhangi Khadtare, R. Sandesh Jadkar, K.N. Hui, S. Rajaram Mane, M. Habib Pathan,. Eosin-Y (EY), rose bengal (RB) and EY-RB sensitized ZnO solar cells (2015) DOI: https://doi.org/10.1109/IRSEC.2014.7059876.
12. S. Sathyajothi, R. Jayavel, A. Clara Dhanemozhi, The fabrication of natural dye sensitized solar cell (Dssc) based on TiO2 using Henna and Beetroot dye extracts, Materials Today: Proceedings **4**, 668–676 (2017)
13. N.A. Al-Omair, S.M. Reda, F.M. Al-Hajri, Effect of organic dye on the photovoltaic performance of dye-sensitized ZnO solar cell. Advances in Nanoparticles **3**, 31–35 (2014)
14. S.S. Kulkarni, S.S. Hussaini, G.A. Bodkhe, D.S. Mahendra, Natural Hibiscus dye and synthetic organic Eosin Y dye sensitized solar cells using titanium dioxide nanoparticles photo anode: comparative study, Surface Review and Letters 1850164 (6 pages). https://doi.org/10.1142/S0218625X18501640