

A novel approach to dye-sensitized solar cells using natural cocktail photosensitizers with nano TiO₂ semiconductor

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Received: 19 March 2021 / Accepted: 20 September 2022 / Published online: 22 October 2022 © The Author(s), under exclusive licence to Springer Science+Business Media, LLC, part of Springer Nature 2022

Abstract

The domain of dye-sensitized solar cells, which use natural dye as a sensitizer, is still being studied. This work aims to determine the most potent sensitizers, namely Geneus Tectona grandis, Chassalia curviflora, and Lawsonia inermis for DSSC fabrication. These dyes were co-sensitized in equal proportions, and the performance of DSSCs was improved through co-sensitization of the natural dyes. The effect of sensitizers and their novel cocktail combination in various solvents namely ethanol and deionized water on DSSC performance is examined. Optical properties and the presence of anchoring groups (carboxyl and hydroxyl groups) in the dye extracts are revealed using UV-Vis and FTIR spectroscopy. Successful accomplishment in the conversion of sunlight into electricity and nano TiO₂-based solar cells using Geneus Tectona grandis, Chassalia curviflora, Lawsonia inermis, and a cocktail of extracts in which Tectona grandis in ethanol displayed the highest photoconversion efficiency of 0.7% compared to co-sensitized dye extracts, whose power conversion efficiency is 0.2%, respectively. The presence of anthocyanin derivative in teak extracts is very sensitive to visible light and thus leads to higher photosensitized performance. Sensitizers for DSSCs must meet crucial parameters such as strong chelation to the semiconductor oxide surface and absorption in the visible and near-infrared portions of the solar spectrum.

Keywords Geneus *Tectona grandis* · *Chassalia curviflora* · *Lawsonia inermis* · Derivatives · Optoelectronics · Conversion efficiency

1 Introduction

Carbon-rich fossil fuels namely natural gas, coal, petroleum, and crude oil were used as the main source of energy all over the world (Richhariya et al. 2017). These resources run out for meeting various needs and liberating poisonous pollutants like sulphur dioxide,

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nitrogen oxides, and carbon emissions contributing to air pollution and global warming (Hafez et al. 2018). These non- recurrent energy sources will be exhausted in the forthcoming years (Coburn et al. 2001). The quest for earth harmony living has stimulated the evolution of substitute technologies that took the edge off harmful pollutants (Kumara et al. 2017). As a consequence, the growth of green technology may be directed toward renewable energy sources for a robust environment (Hafez et al. 2018). The primary sources of renewable energy are solar, wind, hydro, biomass, and geothermal energy. The energy exigency of the future can be achieved by plenitudinous supply, suitable harvesting, and the usage of solar energy (Muhammad Zahir Iqbal 2019). For the clean and sustainable development of the global energy sector, the substitution of fossil fuels with renewable ones has become urgently necessary (Hafez et al. 2018). While collating the properties of various sustainable energy sources, it is so evident that the massive flux of light energy from an average star, the sun can produce a huge amount of energy up to 120KTW approximately, and is far better than the rest of renewable energy technologies having an average output of 5KTW. The Earth receives a tremendous supply of solar radiation from an average star that has been burning for over 4 billion years. Energies stored in all fossil fuel sources is identical to the amount of solar energy that strikes the surface of earth over a 3 day period. This solar energy is limited to 3.28 square meters in India alone. Over 300 days a year, or 5 kW/ m^2 /day on average, of solar energy fall on this land; in some locations, there may be more bright sunny days. It is possible to produce 492,109 kWh/year of electricity at a ten percent overall efficiency even if only 1% of this land is used to harness solar radiation (Coburn et al. 2001). The market for renewable energy in India is now growing more swiftly than the nation's overall energy sector. This enormous amount of energy can be produced using thermal or photovoltaic techniques. India uses both thermal and photovoltaic methods to harness solar energy. Researchers, scientists, and engineers were pursuing the utilization of electricity generation directly from solar light which exhibits the way for solar cell technology through economic devices (Richhariya et al. 2017). With the implementation of photovoltaic cells, the photovoltaic effect generates electric current or voltage when it is exposed to sunlight (Boyle 2004).

Solar cell is hopefully a promising sustainable energy resource that generates electricity with a specified wavelength and wide potential to contribute notably to figuring out the energy crisis faced by future generations. Solar cells are categorized into three generations up to current years. Chronologically, a propitious alternative to replacing waferbased silicon solar cells and thin film solar cells were Dye-Sensitized Solar Cells which are promising PV devices under third-generation solar cells for energy production and are comparatively cheaper. DSSC developed by Michael Gratzel and Brian O' Regan is a novel low-priced chemical photovoltaic cell by the effective combination of nano-crystalline electrodes and charge injection sensitizers. DSSCs are PV devices where visible light into electricity conversion materializes through the sensitization of wide band gap semiconductors (Sathyajothi et al. 2017). It furnishes an economically and technically reliable alternative notion to conventional silicon solar cells or thin film solar cells because of its low efficiency and high cost. Dye-sensitized solar cells is one of the promising and incipient one owing to their innocuous and natural abundance (Richhariya et al. 2017). DSSCs have distinctive features when compared to other solar cells, aside from being low cost in fabrication, made translucent, more flexible, having diverse colors, capable of performing even under scattered light conditions (Kumara et al. 2017), and eco-friendly (Hamadanian et al. 2014).

This novel solar cell technology imitates photosynthesis occurring in plants by converting sunlight into chemical energy by the sensitization of nanostructured TiO_2 substrate

with the help of promising sensitizers Polypyridyl compounds of Ru (II). Howbeit, stability and low conversion efficiency are the crucial complications challenging it. In enhancing the stability of fabricated DSSC and device performance, different light generating systems are implemented to optimize PV performance. The presence of dye sensitizers in the vicinage of nanocrystalline TiO_2 can absorb more energy photons (Eli et al. 2016). Archetypal DSSCs are structurally uncomplicated and composed of a transparent conducting that exhibits an indispensable vast surface area for absorption of dye molecules. A mesoporous semiconductor layer of wide band gap exhibits an indispensable vast surface area for absorption of dye molecules coated on transparent conductive glass. For generating lightstimulated charge carriers, a monolayer of dye sensitizer is attached to the semiconductor surface which forms the photoanode and results in the photosensitizer oxidation. Dye sensitizers are the most salient component for the advancement of the light absorption efficiency of a DSSC. These oxidized charge mediator ions during a reduction in electrolyte scattered towards the counter cathode and receive electrons from the external load. By coupling the photoanode with a counter cathode using a hole conductor ascertain the photovoltaic property of fabricated DSSC. The ability of a dye sensitizer to bind strongly with the nanocrystalline TiO2 semiconductor via anchoring groups namely hydroxyl and carboxyl groups enhances the electron injected into the conduction band of the TiO₂ semiconductor. The current generated by the cell fabricated depends on the vibrant distance betwixt HOMO and LUMO levels of sensitizers. The voltage generated by radiance corresponds to the difference between the Fermi electron level in the nanocrystalline TiO₂ semiconductor and the potential level of the robust electrolyte. The stability and future prospects of solar cells based on novel natural dye mixtures have been addressed in this report (Shalini et al. 2015).

Satria Arief WB et al. synthesized DSSC by using Tectona grandis and obtained a conversion efficiency of 0.0018% (Satria Arief and Yasuri xxxx). Hnin Ei Maung et al. fabricated DSSC by using Lawsonia Inermis and attained a power efficiency of about 0.05% (Hnin Ei Maung 2017). However, in this study, the solar cell was fabricated using Tectona grandis, Chassalia curviflora, and Lawsonia inermis in definite and equal proportions (Syafinar et al. 2015). The performance of fabricated DSSC can be improved by the mixing of natural dye sensitizers as it is an excellent alternative in increasing band absorption. The teak extract was successfully dye-sensitized over a layer of TiO₂ nanoparticles, providing useful information for further studies related to the use of natural pigments as sensitizers for solar cells. Another different perspective put forward by this study is that a cocktail of three individual natural sensitizers in ethanol showed an efficiency of around 0.2%, which is comparatively lower than a single candidate dye. The dye's absorption spectrum and adhesion to the semiconductor's surface have the biggest effects on the solar cell's efficiency. To the best of my knowledge, the cocktail of these three natural extracts is being revealed for the very first time as potential sensitizers for nano TiO₂-based DSSCs and is shown in Fig. 1.



Fig. 1 Natural sensitizers a Lawsonia inermis b Tectona grandis c Chassalia curviflora

2 Experiment

2.1 Preparation of photoanode

2.1.1 Materials and TiO₂ preparation methods

Components include Transparent conducting oxide (FTO), Nano TiO₂ powder, Acetic acid (CH₃COOH), Nitric acid (HNO₃), Deionized water (DI), Ethanol (C₂H₅OH), natural potent sensitizer, Iodine (I), Lithium iodide (LiI), acetonitrile and graphite (Maurya et al. 2016).

0.3 g of colloidal nano TiO₂ is melded with 5 ml of HNO₃ and stirred for 15 min. Every 2 ml of acetic acid is gradually introduced until a thick lump-free homogeneous nanoporous TiO₂ paste is prepared (Maurya et al. 2016). Fluorine-doped tin oxide substrates were sonicated using various solvents namely acetone, ethanol, and deionized water for 30 min in a sonicator bath.

2.1.2 Extraction of pigments

Fresh petals of *Tectona grandis*, *Lawsonia inermis*, and *Chassalia curviflora* fruits were washed thoroughly in solvents namely ethanol and deionized water. Each 10 g of the natural plant sample was dried in a vacuum at 50 °C and then crushed into fine particles using mortar and pestle for 10–15 min. Crushed individual extracts and their cocktail were immersed in 15 ml of solvents separately and kept for a day. Afterward, solid remnants were filtered out and the left-out dye solution was secured from direct solar light and hence used as a potential sensitizer (Maurya et al. 2016). Figure 2, shows the extraction of pigments from natural sensitizers.

2.1.3 Doctor-blade technique

The thickness of sonicated FTO substrate is sealed by scotch tape applied on three ends of the glass film. The aggregate-free nonporous TiO_2 paste is coated on the FTO glass substrate and spread out using the spacer, till a uniform composition is achieved. Before annealing, the nano TiO_2 paste coated on the FTO substrate was let to dry and after drying the paste on a glass substrate, the scotch tape was removed. Using a muffle furnace, the film substrate is then annealed for 450 °C in 30 min to obtain nano-structured



Fig. 2 Extraction of pigments from a Lawsonia inermis b Tectona grandis c Chassalia curviflora

semiconductor TiO_2 film. After annealing, the coated substrate has been cooled for 15 min at room temperature, before immersing the glass film in the dye sample for a week (Maurya et al. 2016). Thus, nano TiO_2 coated film substrate immersed in dye solution forms the photoanode.

2.2 Electrolyte and counter cathode

Robust electrolyte solution employs 0.67 g of Lithium iodide is dissolved in 15 ml of acetonitrile and stirred for 15 min using a magnetic stirrer. 0.13 g of Iodine balls is also added to the solution and then stirred for 10 min till the balls are thoroughly dissolved. The redox couple electrolyte is injected into the unsealed end of the coated film substrate which regenerates the potent dye sensitizers. For counter electrode preparation, nano TiO₂ coated FTO glass film need to be depicted using platinum as the catalyst which can regenerate the redox electrolyte solution. As the platinum catalyst is expensive, a pencil having graphite content is used as a catalyst for a counter electrode.

2.3 DSSC assembling

Finally, the substrate is removed from the dye solution and the sealant is developed on nano TiO_2 coated film substrate to avoid electric contact. For sandwich-type DSSC fabrication, a thin film photoelectrode is put on top of the counter electrode where the conductive end of the counter cathode can face the FTO substrate coated with dye sensitizers. Thus, the DSSC cells were binded using crocodile clips and sandwiched together for measuring electrical performance. By capillary action, the redox electrolyte solution was injected into the space between the glass films (Maurya et al. 2016). Figure 3, displays the photo-electrochemical studies taken for determining the J-V characteristics of the fabricated DSSC.

3 Results and discussion

3.1 Absorption spectra of sensitizers

UV-Vis Spectrophotometer is used for analyzing the optical spectrum of solvents namely ethanol and deionized water attained from extracts of *Tectona grandis*, fruits of



Fig. 3 Photo-electrochemical Studies



Fig. 4 UV-Vis absorption spectra of Lawsonia inermis in ethanol and DI



Fig. 5 UV-Vis absorption spectra of Tectona grandis in ethanol and DI



Fig. 6 UV- Vis absorption spectra of Chassalia curviflora in ethanol and DI



Fig. 7 UV-Vis absorption spectra of Cocktail dyes in ethanol and DI

Chassalia curviflora, and *Lawsonia inermis*, absorbed into nanoporous TiO_2 coated surface is depicted in Figs. 4, 5, 6 and 7. The efficiency of the cell is strongly influenced by the absorption spectrum of the dyes and the injection of electrons into the surface of the thin nano-crystalline layer of TiO_2 by excited sensitizers. Natural derivative-rich dye sensitizers were used for the synthesis of dye-treated titanium dioxide nano films which yield colored nano substrates with high absorption in the visible range. The dye sensitizers indicate absorption in the range of 400–700 nm with peaks observed at 455 nm for *Tectona grandis* which can be attributed to the dye pigments present in the plant extracts. From these absorption graphs, it is obvious that these dye extracts absorb light in the visible region and therefore satisfy the primary canon as natural sensitizers in the fabrication of solar cells. *Tectona grandis* contains anthocyanin pigment, which is the core component of dye sensitizers and is commonly found in leaves, fruits, and flowers of plants. It is anticipated that its red color will make nanostructured TiO_2 an efficient dye sensitizer (Maurya et al. 2016; Ananth et al. 2014). The HOMO and LUMO levels of anthocyanin

are thought to be impacted by the anthocyanin-containing dye's attachment to TiO_2 , which in turn reduces the band gap and causes the absorption peak to shift to the red. Besides, owing to the presence of binding groups namely hydroxyl and carbonyl, molecules in anthocyanin derivative can bound easily with TiO_2 substrate, which enhances its application as a dye sensitizer. Tannins and their derivatives in *Chassalia curviflora* are polyphone chemicals that can create insoluble colored compounds through strong complexation activity with Ti⁴⁺. Tannins can therefore be used as an effective sensitizer. Chlorophyll, which is able to produce glucose from simple organic molecules (water and carbon dioxide) when exposed to visible light, are present in *Lawsonia inermis*. As a result of their capacity to absorb photons from sunlight and their ability to collect light from the infrared spectrum, chlorophyll pigments can be employed as a dye sensitizer in DSSC (Syafinar et al. 2015). It indicates that natural extracts are thoroughly adsorbed onto the surface of wide band gap TiO₂ semiconductor which may rise the light absorption by the particles of semiconductor oxide in the visible range owing to the binding of derivatives present in the dye samples to the TiO₂ surface (Maurya et al. 2016). The wavelength range absorbed is connected to the band gap, which is the photon energy of TiO₂, and the band gap shrinks as the absorption wavelength increases. The dye's lowest band gap facilitates the electron's quick transition from the valence band to the conduction band, requires less energy for electron recombination, and yields a high fill factor (Syafinar et al. 2015).

The variations and differences in the absorption properties of natural sensitizers can be ascribed to the various colors of the dye extracts owing to their respective derivatives in them. Hence, the absorption peaks attained by the potent dye sensitizers in solvents namely ethanol and deionized water, and bandgap energy calculated from the Tauc plot for the natural dyes are depicted in Table 1. The bandgap energy of the sensitizers is calculated using the equation given below.

$$E_{g} = \frac{hC}{\lambda e}$$
(1)

3.2 Fourier transform infrared spectroscopy

FTIR spectra are used to evince the presence of anchoring groups especially (O–H), (COOH), (C=O), and (=O) available in the potential dye which proves the compatibility of the sensitizer with the nano TiO_2 surface. Phytochemicals can bind with the TiO_2

Serial No	Sensitizers	Ethanol	Deionized water		
		λ _{max} (nm)	Bandgap energy (eV)	λ_{max} (nm)	Bandgap energy (eV)
1	Tectona grandis	455 nm	2.57 eV	404 nm	2.72 eV
2	Chassalia curviflora	402 nm, 404 nm, 481 nm	2.81 eV	470 nm, 494 nm	2.40 eV
3	Lawsonia inermis	551 nm, 555.5 nm 625 nm	1.87 eV	404 nm, 408 nm,413 nm, 439 nm 475 nm	2.44 eV
4	Cocktail dyes	452 nm, 523 nm	2.22 eV	400.5 nm	2.70 eV

Table 1 depicts the optical absorption range and bandgap energy of extracted natural dye sensitizers

semiconductor and have an impact on the color of plant materials. The infrared spectrum of natural sensitizers was recorded in the range of 4000-400 cm⁻¹ and displayed in Fig (Hamadanian et al. 2014; Eli et al. 2016; Shalini et al. 2015; Satria Arief and Yasuri xxxx). An inspection of the Tectona grandis spectrum exhibits that they reveal broad absorption ranging from 3000 to 3700 cm^{-1} with a strong band at 3340.71 cm^{-1} and 3271.27 cm⁻¹ which is ascribed to the compound class alcohol having O-H stretching group and owing to the large variety of hydroxyl linkage (Maurya et al. 2016). It has a sharp peak at 2978.09 cm⁻¹, 2885.51 cm⁻¹, 2762.06 cm⁻¹ allocated to C=O stretching vibrations, and a medium peak at about 1381.03 cm⁻¹ due to O-H bending vibration mode. The cocktail of dyes namely Tectona grandis, Lawsonia inermis, and Chassalia *curviflora* in ethanol exhibit broad and strong peaks at 3379.29 cm⁻¹ and 3302.13 cm⁻¹ which is attributed to hydrogen bonding between O-H stretching alcohol group and broad peak at about 2978.09 cm⁻¹, 2769.78 cm⁻¹, 2638.62 cm⁻¹, 2561.47 cm⁻¹, 2515.18 cm⁻¹ owing to C=O linkage. The spectrum also reveals a medium peak at 1365.60 cm⁻¹. Likewise cocktail of dyes in deionized water also exhibits a broad peak at 3379.29 cm^{-1} , 3016.67 cm⁻¹ having O-H vibration mode, and a strong, wide peak at 2970.38 cm⁻¹, 2831.50 cm⁻¹, 2654.05 cm⁻¹, 2561.47 cm⁻¹ ascribed to the carbonyl group. The anchoring groups confirm the presence of anthocyanin, Tannin, and chlorophyll pigments. The disappearance and shifting of peaks in the FTIR spectrum are caused by the dilution of salts with polar solvents, which weaken the bonds in the molecular configurations. Phytochemicals present in dye extracts may be responsible for the observed functional groups. The phytochemicals present in *Tectona grandis* are triterpenoids, anthraquinones, isoprenoid quinones, phenolic compounds, naphthoquinones, lignans, fatty esters, and steroids (Vyas et al. 2019). Chassalia curviflora extracts include carbohydrates, triterpenoids, tannins, alkaloids, phenolic compounds, saponins, flavonoids, glycosides, and phyto-steroids whereas Lawsonia inermis includes the presence of tannins, anthraquinones, flavonoids,



Fig. 8 FTIR Spectra of Lawsonia inermis in ethanol and DI



Fig. 9 FTIR Spectra of Tectona grandis in ethanol and DI



Fig. 10 FTIR Spectra of Chassalia curviflora in ethanol and DI



Fig. 11 FTIR Spectra of Cocktail dye in ethanol and DI

saponins, alkaloids, phenol, and glycoside FTIR spectra of sensitizers in ethanol and deionized water are depicted in Figs. 8, 9, 10, 11.

3.3 J-V characterization

To measure the photovoltaic performance of fabricated sandwich structured DSSC with natural potent dyes as effective sensitizers, using Peccell Solar Simulator having irradiance around 100 mW/cm² to stimulate solar light. Under the illumination of visible light, the current–potential curve of constructed DSSC with dye-capped nano TiO₂ and counter cathode can be recorded. The maximum energy conversion efficiency can be studied using the J–V curve which determines the operational parameters such as open-circuit voltage (Voc), short-circuit current density (Jsc), Fill factor (FF), and conversion efficiency is calculated using the equation,

$$FF = (J_m \cdot V_m) / (J_{sc} \cdot V_{oc})$$
⁽²⁾

$$\eta\% = \frac{FF.J_{sc}.V_{oc}}{Input \ power}.100$$
(3)

The J–V characteristics of the fabricated solar cells are depicted in Figs. 12, 13, 14 and 15. The performance of DSSC using *Tectona grandis* in ethanol reported higher power conversion efficiency of around 0.7% than the previous records. *Lawsonia inermis* in deionized water and co-sensitized extracts in ethanol showed an efficiency of about 0.2%. Hence, *Tectona grandis* in ethanol is an effective candidate for stable DSSC. In a relatively



Fig. 12 J-V curve of Lawsonia inermis in ethanol and DI



Fig. 13 J-V curve of Tectona grandis in ethanol and DI

short period of time, the anthocyanin in *Tectona grandis* exhibits a strong affinity for TiO_2 nanoparticles and improves their performance. The ability of the photocurrent to produce energy from incident light grew along with the concentration. It was also reported that the degradation of the solar cell performance is due to the detachment of the dye from the TiO₂ surface as depicted in Figs. 16, 17. The variation in sensitization actions of dyes that take place on different semiconductor surfaces causes variations in photovoltaic performance. This is caused by a variation in how dye molecules transmit electrons to semiconductor conductor conduction bands. In semiconductor films, impure dye causes dye aggregation,



Fig. 14 J-V curve of Chassalia curviflora in ethanol and DI



Fig. 15 J-V curve of Cocktail dyes in ethanol and DI

which interferes with light absorption. The conversion efficiency attained by the sensitizers in solvents namely ethanol and deionized water is depicted in Table 2.

4 Conclusion

Natural dyes contain a variety of pigments, including anthocyanin, tannin, and chlorophyll, which can absorb photons from sunlight and convert them to electrical energy. The natural dyes extracted from *Tectona grandis*, *Chassalia curviflora*, *Lawsonia inermis*, and a novel cocktail combination of These dyes were introduced as a promising sensitizer for effective



Fig. 16 Photo-sensitization performance of DSSC in Ethanol



Fig. 17 Photo-sensitization performance of DSSC in Deionized water

fabrication of DSSC. The extracted dyes of *Tectona grandis* contain natural anthocyanin pigment, *Chassalia curviflora* fruit has a tannin derivative, and *Lawsonia inermis* contains chlorophyll derivative, which is responsible for imparting color to the TiO₂-based photo-anode. UV–Vis Spectra determine maximum absorption possessed by the dyes in the visible range. The DSSC sensitized by *Tectona grandis* offered the highest efficiency of about

Serial No	Sensitizer	V _{oc}	J_{sc} (mA/cm ²)	V_m	$J_m (\mathrm{mA/cm^2})$	FF	η %
Ethanol							
1	Tectona grandis	0.43	5.21	0.32	2.16	0.3	0.7
2	Chassalia curviflora	0.2	0.78	0.122	0.3	0.2	0.03
3	Lawsonia inermis	0.3	0.81	0.2	0.5	0.4	0.1
4	Cocktail dyes	0.24	2.6	0.17	1.0	0.3	0.2
Deionized v	vater						
1	Tectona grandis	0.06	0.33	0.03	0.13	0.2	0.004
2	Chassalia curviflora	0.22	1.44	0.17	0.53	0.3	0.1
3	Lawsonia inermis	0.27	3.5	0.18	0.94	0.2	0.2
4	Cocktail dyes	0.3	1.4	0.21	0.63	0.3	0.13

 Table 2 Depicts the conversion efficiency attained by the sensitizers

0.7% than the previous record. A new cocktail combination of *Lawsonia inerrmis*, *Tec*tona grandis, and *Chassalia inermis* dyes showed an efficiency of around 0.2% which is comparatively lower than single candidate dyes. The study report concludes that the influence of solvents, namely ethanol, also plays an indispensable role in solar cell performance than deionized water because higher polarity solvents enable the sensitizers to adsorb more energy, which leads to faster electron injection and more effective solar cell technology. The power conversion efficiency is expected to be ameliorated by introducing anchoring groups namely (–OH) and (–COOH) to the sensitizers. Future efforts will be made to increase the effectiveness of natural dyes by optimizing DSSC performance for glass plate thickness and sample coating area on FTO glass substrate.

Author contributions PP: Conceptualization; Investigation; Data curation; Formal analysis; Writing—original draft. NR: Investigation; Resources. SS: Investigation; Resources. BJ: Validation; Supervision.

Data availability The datasets generated during and/or analyzed during the current study are available from the corresponding author upon reasonable request.

Declarations

Conflict of interest None to declare.

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