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Locally Available Natural Dyes for Dyesensitized Solar Cells

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ABSTRACT

Different types of natural dyes are commonly used to fabricate dyesensitized solar cells which are cheaper, simpler, and environmentally friendlier than conventional crystalline silicon solar cells. This paper objectifies the performance of locally grown natural dyes from Dhaka, Bangladesh using a simple fabrication design. Four natural dyes were obtained separately from spinach, turmeric, pomegranate, and beetroot which contain chlorophyll, curcumin, anthocyanin, and betanin pigments respectively. Ultraviolet-visible spectroscopy illustrated the optical properties of these dyes diluted in ethanol. Spinach and turmeric extracts showed sharper absorption peaks which qualify them as better sensitizers than pomegranate and beetroot extracts. Photoanodes (of thickness 15-20 µm) were prepared by employing the doctor blade technique on fluorine-doped tin oxides using TiO_2 nanoparticles of anatase form and graphite coated fluorine-doped tin oxides were used as counter electrodes. The photoelectric measurements of the fabricated cells were done under air mass (AM) 1.5 and power density of 100 mW/cm^2. The fabricated cell of turmeric extracted dye showed the highest efficiency of 0.031% (Open circuit voltage, V_{OC} = 380 mV, Short circuit current density, J_{SC} = 0.234 mA/cm^2) and the cell using beetroot extract gave the highest fill factor (FF) of about 50% among the prepared dyesensitized solar cells.

Keywords: DSSC, Gratzel cells, FTO, Low-Cost, Natural dye.

1. Introduction

Dye sensitized solar cells (DSSCs) are potential candidates to replace regular silicon-based solar cells because of their low manufacturing cost, easy fabrication process, and eco-friendliness [1-2]. These cells are also known as Gratzel cells and were originally co-invented by Brian O'Regan and Michael Grätzel at UC Berkeley in 1988 [3]. A dyesensitized solar cell is primarily composed of a nanoporous TiO_2 coated photoanode, a dye monolayer, a redox electrolyte, and a counter electrode. Nevertheless, various traditional and novel materials are being employed by researchers to change the performance and stability of these cells [4]. Due to its crucial part in DSSCs, a significant amount of research is being performed on the improvement and selection of dyes [5]. When first reported in 1991, ruthenium polypyridyl complexes had been used as a sensitizer because of their various advantages such as intense charge transferring capability, presence of effective anchoring group, etc. [6]. But Ruthenium based organic dyes are costly and chemically toxic which led to exploration into natural dyes extracted from fruits, vegetables, etc. [7]. The highest efficiency of 14.30% has been reported using metal-free organic dyes [8].

The plant pigments found in the most common sources of natural dyes can be classified into four major categories- (i) Tetrapyrroles, (e.g. green chlorophylls); (ii) Carotenoids, (such as yellow curcumin); (iii) Flavonoids, (red, purple or blue anthocyanins), and (iv) Betalains (e.g. betaxanthins, betacyanins, etc.) [9]. Chlorophylls (Chl) are the most abundant natural pigment which can absorb solar energy in different regions of the visible spectrum [10]. There are several types of chlorophylls among which, Chl-a and Chl-b are two major types [11]. Carotenoids are also prolifically found in nature. Curcumin is a xanthophyll carotenoid and has plenty of uses (most common of which is as a spice) in East Asian countries [12, 13]. Anthocyanins are the reason behind the coloration of various plants and readily bind with TiO_2 surface due to the presence of carbonyl and hydroxyl groups [14]. Betalains have a colored compound known as betalamic acid. Betalains have two types- (a) red-purple betacyanins and orange-yellow betaxanthins [15]. The most common betacyanin and betaxanthinarebetanin (red-colored) andindicaxanthin (yellow colored) respectively [16, 17].

In the present work, chlorophyll dye was extracted from spinach (Spinacia oleracea), curcumin dye from turmeric (Curcuma longa), anthocyanin dye from pomegranate (Punica granatum), and betanin dye from red beetroot (Beta vulgaris) using ethanol in a 1:10 ratio. Next, the optical properties of the extracts and the structural property of TiO_2 thin films were investigated. Using the extracted dyes, solar cells were assembled applying a very cheap and simple fabrication method. Finally, electrical characterization was done to evaluate performances of the DSSCs and subsequently, the best solar cell using natural pigment as sensitizer was determined.

2. Experimental Details

2.1. Chemicals & Materials

Fluorine-doped tin oxide (FTO) glass (sheet resistance < 7 Ω/cm^2), TiO_2 nanopowder, distilled water, 69% HNO_3, 0.5 M Lithium Iodide (LiI), 0.05 M Iodine (I), and Acetonitrile (ACN) were used during the cell preparation. All chemicals and solvents were provided by the thin-film solar cell laboratory of the Institute of Fuel Research and Development (IFRD), Bangladesh Council of Scientific & Industrial Research (BCSIR). All the purchased chemicals and solvents were used without further purification.

2.2. Dye Extraction

Freshly plucked pomegranate, beetroot, spinach, and turmeric were used to extract natural dyes. All of these vegetables and fruits were locally grown and collected directly around different locations in Dhaka, Bangladesh. These items were cleaned with distilled water before extracting natural dye from them. Beetroot, spinach, and turmeric were cut into small pieces. The arils of pomegranate were separated from its peel and pulp membrane. Then those arils were crushed by hands, seeds were removed from the juice. Ethanol was mixed into the raw spinach, turmeric, pomegranate juice, and beetroot separately in a1:10 ratio and kept in darkness overnight. The subsequent extracts were filtered to remove solid fibers or fragments. Then a portion of the ethanol extracts was used for Ultraviolet-visible (UV-vis) characterization and the remaining dyes were reserved at room temperature under dark condition.

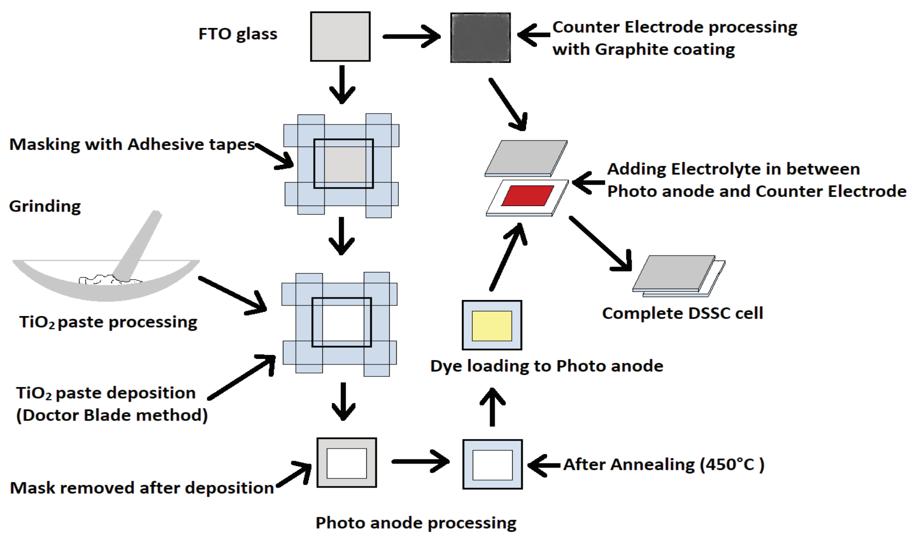

Fig. 1: DSSC Fabrication Process

2.3. Preparation of Photo anode

2×2 cm sized FTO glasses were cleaned using an ultrasonic bath in ethanol, acetone, and distilled water for a total of 30 minutes (10 minutes each step). Purchased nano-crystalline TiO_2 powder and other chemicals have been used without any further purification. At first, 2 gm of TiO_2 powder was ground in an agate mortar with a pestle for 1hour. Then, 0.9 ml of HNO_3 was dropped into the ground powder and was continuously mixed by a pestle for about 3-4 minutes until a slurry paste was obtained. Doctor blade method was employed to deposit TiO2 paste on cleaned FTOs. Then the edges of the conductive side of the substrates were covered by adhesive tapes to mask electric contact strips and to control film thickness. The TiO2 paste was then uniformly rolled over by a glass rod and kept at room temperature for 20 minutes. To improve crystallinity, the prepared electrodes were annealed at 450°C for 30 minutes and then gradually cooled down to room temperature. X-ray diffraction was carried out on these annealed films to determine the crystallinity of the films. The subsequent film thickness is around 15-20 µm, measured by a surface profilometer. Next, the thin films were immersed in the filtered dye to sensitize the semiconductor materials. Fig. 1 represents the entire DSSC fabrication process.

2.4. Electrolyte and counter electrode preparation

Solution of 0.5 M Lithium Iodide (LiI), 0.05 M Iodine (I), 2 ml Acetonitrile (ACN) was used as Redox (I^-/I^{3-}) Electrolyte Solution. The conductive side of FTOs was coated by a 10B pencil. The loosely attached large particles were carefully removed before using the coated FTOs as counter electrodes.

2.5. Cell Fabrication

After 24 hours of soaking in dark conditions, the dye absorbed films were rinsed in distilled water to detach the unbound particles and dried in the air. The working anode was then ready to use in cell fabrication. The photoanode and the counter electrode were clamped together in a sandwich formation so that the absorbed dye and the coated graphite could face each other. They are done so in a slightly offset manner so that electric contact could be allowed later. 1 or 2 drops of redox electrolyte were injected between the electrodes such that the total intersected area was covered. Using the same method and materials described above, four different DSSCs were fabricated. These cells are only distinguishable by their respective sensitizers (spinach, turmeric, pomegranate, and beetroot).

Cell name	Source dye	pigments
DSSC-1	Spinach	Chlorophyll
DSSC-2	Turmeric	Curcumin
DSSC-3	Pomegranate	Anthocyanin
DSSC-4	Red beetroot	Betanin+indicaxanthin

Table-1: List of fabricated DSSCs using different dyes and their pigments

2.6. Characterization and measurement

The absorption spectrum of the samples was determined using a UV-vis spectrophotometer (Cintra 2020, GBC). X-ray diffraction (EMMA, GBC, Australia) with Cu K-α radiation (λ = 0.154 nm) was used to identify the crystallinity of the TiO_2 films. Film thickness was measured using a Surface Profilometer (Dektak XT-A, Bruker). The photovoltaic tests were carried out by Sun Simulator (K3000LAB55, Mcscience) in the ambient atmosphere. The energy gap of dye was determined by using the formula of equation (1). Here, E is the photon energy or optical energy gap.

$$E = h\nu = (hc) / \lambda \quad (1)$$

Where, h = Planck's constant (6.63×10^{-34}Js), ν = Frequency, λ = Wavelength, c = Speed of light (3.0×10^{8} m/s). The absorption coefficient (α_{abs}) was obtained as follows:

$$\alpha_{abs} = (4\pi K) / \lambda \quad (2)$$

Where K is extinction co-efficient. To calculate the average crystallite size (D) of the TiO_2 nanoparticles, the Scherrer formula was used,

$$D= (k\lambda_{x\text{-}ray}) / (\beta \cos\theta) \quad (3)$$

Where k = Scherrer constant (0.9), $\lambda_{x\text{-}ray}$ = wavelength of X-ray used ($\lambda_{x\text{-}ray}$ = 0.15406 nm), β = full width at half maximum (FWHM) and θ = scattering angle.

3. Result And Discussion

3.1. UV-vis spectroscopy

3.1.1. Absorption spectra analysis

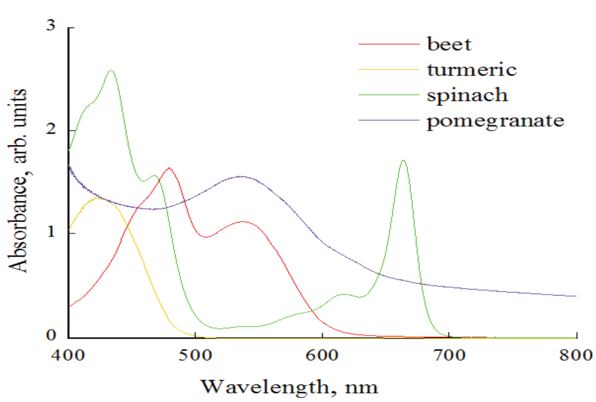

Fig. 2: UV-vis absorption spectra of beetroot, turmeric, spinach, and pomegranate extracted dyes

Broader absorption spectra and higher absorption intensity in the visible spectrum are desirable in natural dyes [14, 18]. The optical properties of the extracts were analyzed using UV- vis spectroscopy. Fig. 2 presents the UV-vis absorption spectra of beetroot, turmeric, spinach, and pomegranate extracts with ethanol in the visible spectral range (380-800 nm).

Spinach extract exhibits absorption peaks at 435 nm and 470 nm with a sharp rise at 664 nm in the absorption range of 600-700 nm. These spectral responses show the presence of Chlorophyll pigment [19-21]. Optical absorption of turmeric extracted dye shows a climb at λ_{max}(wavelength where peak obtained) = 422 nm in the 380-500 nm region which attributes the presence of curcumin [22, 23]. For pomegranate extracted dye, the absorbance peak (λ_{max}= 535 nm) occurs in the 500-600 nm region and signifies the presence of anthocyanin pigments [24-26]. Beetroot extract shows a broad range of absorption from 400 to 600 nm and is expected to be a mixture of several pigments. It has two peaks at 479 nm and at 537 nm which can be attributed to the presence of betanin and indicaxanthin respectively [16, 27-28]. From UV-vis spectra, it can be seen that pomegranate and beetroot extracted dyes have flatter peaks than other extracts. This behavior of the dyes indicates that they are poorer sensitizers than turmeric and spinach extracts [29].

3.1.2. Band gap estimation and absorption coefficient of the natural dyes

The light absorption capacity of the dye is an important factor considering the improvement of cell performance [30]. The energy gap of a dye is the difference between the lowest unoccupied molecular orbital (LUMO) and highest occupied molecular orbital (HOMO) levels. Dyes with a narrow band gap captures low-energy photons. On the other hand, the absorption coefficient is a function of color and measures the amount of absorbed light for a fixed thickness of the material. The absorption coefficient characterizes how far into the material light of a particular wavelength can penetrate before it is absorbed [31]. Optical band gaps or optical energy gaps (E) and absorption coefficients (α_{abs}) of all the extracted dyes have been found using equations (1) and (2) respectively and the results are listed in Table-2. Band gaps have been found in the range of 1.87-2.94 eV and α_{abs} has the range of 1.63-2.57 km^{-1}. Turmeric extracted curcumin dye has the largest E (2.94eV) and α_{abs} (2.57 km^{-1}) values while chlorophyll has the lowest respective values (E = 1.87 eV and α_{abs} = 1.63 km^{-1}).Kabir et al. have found peak absorbance at 662 nm (absorption range 600-700 nm) resulting in similar optical bandgap and absorption co-efficient (1.87 eV and 1.57 km^{-1} respectively) [19]. Ruhane et al. found absorption peaks for turmeric at 448 nm (optical band gap 2.77 eV and absorption co-efficient 2.42 km^{-1}) for their ethanol-based samples [32]. María et al. examined different beetroot samples and found peaks at 516 nm, 519 nm, and 526 nm [9]. Mozhgan et al. found absorption peaks for sour and sweet pomegranate at 511 nm and 525 nm respectively [24]. Interestingly, anthocyanin and betanin showed almost identical values of E and α. The optical band gap of a semiconductor material is the difference between the conduction (VB) band and valence band (CB). The calculated band gap for TiO_2 particles is 3.12 eV.

Dye Sources	Absorption Range (nm)	Absorption peak, λmax (nm)	Energy Gap, E (eV)	Absorption Coefficient, α (km-1)
Spinach	600-700	664	1.87	1.63
Turmeric	380-500	422	2.94	2.57
Pomegranate	500-600	535	2.32	2.02
Beetroot	400-600	537	2.31	2.02

Table-2: Energy gaps (E) and absorption coefficients (α) of four extracted dyes.

Zainal et al. compared their natural sensitizer's performance with a commercial dye, N719 and found the absorption peak at 515 nm (calculated optical bandgap 2.41 eV and absorption co-efficient 2.03 km^{-1}) whereas María et al. found absorption peak for N719 at 533 nm (calculated optical bandgap 2.33 eV and absorption co-efficient1.96 km^{-1}) [2, 9].

3.2. X-ray Diffraction (XRD) data analysis

X-ray diffraction technique was performed to analyze and identify phase formation, crystallographic information, and crystallite size of TiO_2 thin films. Fig. 3 shows the XRD patterns in the range of 20-80° of the deposited TiO_2 on FTO after annealing at 450°C. The choice of deposition method can significantly influence the phase formation and crystallinity of the deposited TiO_2 thin films [33].

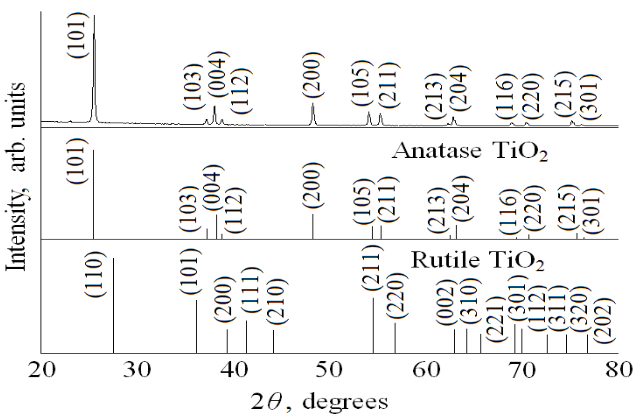

Table-3: Electrical properties of assembled natural dye sensitized solar cells.

All the peaks in Fig. 3 can be indexed as tetragonal TiO_2 anatase phase and are analogous to the peaks found in ICDD. A sharp diffraction pattern at anatase (101) implies a high degree of crystallinity of the anatase phase. Some minuscule peaks which cannot be indexed are due to impurities of purchased TiO_2.The anatase phase has greater photoactivity, band gap, refractive index, and electron diffusion coefficient while the rutile is more thermally stable, and can absorb blue light [34]. Regardless of the exact reasons, studies have shown that the TiO_2 anatase phase is a better photocatalytic agent than the rutile phase [35-37].For this experiment, the crystallite size was found to be 31.7 nm for the anatase (101) peak using equation (3).Smaller crystallite sizes can have a higher surface area that helps dye particles to adhere and as a result can improve cell performance [38, 39].

3.3. Photoelectric properties of DSSCs

The photoelectrical measurements of the four prepared DSSCs were performed under air mass (AM) 1.5 conditions with an incident light power density of 100 mW/cm^2. All the values of V_{OC}, J_{SC}, FF, η, R_{sh}, and R_s were directly found from the sun simulator. Table-3 summarizes the performances of all the fabricated DSSCs and Fig. 4 shows the short circuit current density vs. voltage (J-V) curve of them.

From Table-3, it is evident that DSSC-2 gives the highest V_{OC} of 380 mV which has contributed to its highest efficiency (η = 0.031%) among the fabricated dyes. DSSC-1 shows slightly lower values than DSSC-2 (η = 0.027% and V_{OC} = 371 mV) but has the highest short circuit current density (J_{SC} = 0.240 mA/cm^2) which may be due to the better light absorption capability of chlorophyll than curcumin pigment. Nevertheless, the higher band gap of turmeric extracts may be the reason behind the high V_{OC} value of DSSC-2 which might have contributed to its performance. Even though pomegranate and beetroot had shown similarities in their E and α values, DSSC-3 and DSSC-4 showed variability in their values of V_{OC} (297 mV, 255 mV), J_{SC} (0.235 mA/cm^2, 0.103 mA/cm^2), and η (0.018%, 0.013%). The reason for this may be because of the lack of strong bonding of anthocyanin and betanin and/or indicaxanthin molecules with TiO_2 particles.

Dye Sources	VOC (mV)	JSC (mA / cm²)	Fill Factor, FF (%)	η (%)	RSH (Ω)	RS (Ω)
DSSC-1	371	0.240	30.61	0.027	923.44	833.85
DSSC-2	380	0.234	34.72	0.031	1728.17	542.54
DSSC-3	297	0.235	26.25	0.018	1147.42	1248.45
DSSC-4	255	0.103	49.76	0.013	4618.36	858.38

Fig. 3: XRD pattern for TiO2 coated on FTO surface after annealing in 450 degrees for 30 minutes (top), anatase and rutile.

Evidently, DSSC-4 has a fill factor of ≈50% and the fill factor of DSSC-3 is ≈26%. This large variation of FF values can be described by explaining the distinct values of shunt resistance (R_{SH}) and series resistance (R_S).Higher R_{SH} and lower R_S positively affect the fill factor value i.e. increasing the difference between shunt and series resistance of a particular cell will lead to an increasing filling factor [40]. For DSSC-4, this difference is about 3760 Ω and consequently has the highest filling factor of ≈50%; whereas, for DSSC-3, this value is -101 Ω ($R_S > R_{SH}$) which explains its lowest FF (≈26%).

DSSCs using synthetic dyes can achieve much better efficiency- such as, Zainal et al. used commercial dye- N719 and gained 0.728% efficiency for 1.0 × 10^{-7} mol/cm^{2}dye loading. For N719 based cells in their study,V_{oc} was as much as 732.92 mV and J_{sc} was 2.451 mA/cm^2 [2]. The overall efficiencies found in this work (0.013-0.03%) are lower than the efficiencies reported in the literature [29, 32, 41]. Although the V_{OC} of DSSC-1 (371 mV) is better than reported by Kabir, et al. [41] (336 mV), the efficiency of DSSC-1 (0.027%) is lower than the reported cell (0.398%). This is because the cell in Kabir, et al. had a larger short circuit current (2.435 mA/cm^2) than DSSC-1 has (0.529 mA/cm^2). The low short circuit current value in the present work may have occurred due to the use of ethanol as an extraction solvent rather than methanol or by the employment of different electrolytes used by Kabir, et al. In the case of DSSC-2, the V_{OC} value 380 mV can be compared to the value 428 mV found by Ruhane, et al. [32] but they also have reported a slightly better efficiency (0.05%) than the highest efficiency reported in this work (0.031%). This difference may be attributed to their use of ITO as a substrate instead of FTO. Table-3 shows, DSSC-3 gives a better current density, better voltage as well as better efficiency, than DSSC-4 which supports the conclusion drawn by Kavitha, et al. that the fabricated cells with pomegranate dye show better efficiency than using beetroot extracts. The higher FF value in the beetroot-based cell is also analogous that was due to the lower series resistance in the pomegranate-based cell [29]. Furthermore, the efficiency of DSSC-4 (0.013%) is marginally greater than the value of cell prepared with beetroot dye (0.0119%) by Kavitha, et al. However, the efficiency of DSSC-3 (0.018%) is less than the efficiency of the fabricated cell using pomegranate dye (0.268%) reported by the same author because their cell had a higher V_{OC} (340 mV) and FF (35%) which resulted in higher efficiency (0.268%). The lower V_{OC} (297 mV) and FF (26%) of DSSC-3 might have originated from the fact that the counter electrode used here was graphite coated, contrary to the platinum used in their work.

Stefan et al. achieved 0.028% efficiency with 45% FF for DSSCs stained with turmeric dye [42]. Turmeric includes favorable functional groups that help better adhesion with porous TiO_2 [32, 43]. Choawunklang et al.compared cell efficiencies prepared from different natural dyes (turmeric, saman bark, bai-ya-nang, butterfly pea, and black rice) and found maximum efficiency from turmeric dye despite greater FF values of other dyes. Karim et al. found better efficiency and better cell voltage when they compared turmeric dye-based DSSCs with carissa carandas and beet though FF values of the turmeric-based cell were almost half of the samples from other dyes [44].

4. Conclusion

In this present work natural spinach, turmeric, pomegranate, and beetroot from Dhaka, Bangladesh were selected as the sensitizers for DSSC fabrication. These dyes contain chlorophyll, turmeric, anthocyanin, and betanin pigments respectively. Optical analysis of these dyes showed that spinach and turmeric extracted dyes were better sensitizers than pomegranate and beetroot extracts due to their sharper absorption peaks. Solar cells have been assembled using these natural dyes and TiO_2 nanoparticles of the anatase phase. The electrical measurement of the fabricated DSSC using turmeric extract gives the highest conversion efficiency of 0.031% and V_{OC} of 380 mV following by DSSCs using spinach extract ($\eta = 0.027\%$ and $V_{OC} = 371$ mV). Cells using pomegranate extract ($\eta = 0.018\%$) and beetroot extract ($\eta = 0.013\%$) gave the lowest FF ≈ 26% and the highest FF ≈ 50% respectively. Despite, the low efficiency of the fabricated cells, the method and materials used in this work are handy and simple enough to carry out further research on naturally available dyes in Bangladesh.

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