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Fabrication and Characterization of Dye-Sensitized Solar Cells Using Natural Dyes

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ABSTRACT

This work reflects the fabrication and characterization techniques of Dye-Sensitized Solar Cells (DSSCs). The DSSCs is one of the foremost promising 3rd generation solar cell due to the greener energy production. The goal of this work was to create a low cost TiO_2-based dye-sensitized solar cells (DSSCs) with a carbon counter electrode and natural dyes as sensitizing agent. The extracted dye components were shown a notable absorbance in between 400nm to 700nm wavelength in UV-Vis spectrophotometer. The photoelectrochemical characteristics of the DSSCs were then characterized under AM 1.5 illumination of solar simulator. The open circuit voltages and short circuit currents density were achieved 0.52 V, 0.44V, 0.345V and 0.531 mA/cm^2, 0.267 mA/cm^2, 0.31 mA/cm^2 for *Tradescantia pallida*, *Jungle geranium*, and *Catharanthus roseus* based DSSCs respectively. Whereas the efficiency was 0.095%, 0.0611%, 0.0528% respectively.

1. Introduction

There has been a lot of research into the creation of dye-sensitized solar cells (DSSCs) as an alternative technology for applications involving solar power. Compared to conventional silicon-based cells, DSSCs provide benefits including cheaper manufacturing costs and less harmful materials [1]. The high bandgap semiconductor metallic oxide that has dye molecules adsorbed on it serves as the photoelectrode in DSSC devices. A counter-electrode comprised of a catalyst like platinum covers the conducting glass. An excellent light scattering effect occurs inside the porous photoelectrode as a result of the elevated refractive index of anatase TiO_2, which is crucial for improving light harvesting effectiveness [2]. To aid in electron transport and restore diminished sensitizer, the electrolyte is pumped in between the electrodes. The redox pairs of iodide/triiodide that are dispersed in a liquid organic solvent make up the majority of the electrolytes that are employed [3].

The most effective DSSCs were developed utilizing dyes made of ruthenium bipyridyl complexes, which are highly stable, easily exploited by semiconductor films, and display outstanding electron injection capabilities.

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These dyes absorb a great percentage of solar radiation (primarily visible illumination). M. D. A. Sanda et al. gained cell efficiency of 0.05% and 0.07% using natural dyes [7]. P. Gu et al. achieved a cell efficiency of 0.051% using Mulberry as a sensitizer [8]. R.N. Iman et al. achieved 0.31% of cell efficiency [12]. G. Richhariya et al. found 3.26% of power conversion efficiency [13]. F. Aslan availed 0.47% of power conversion efficiency [14]. A.K. Alanazi found 0.095% of efficiency using green cabbage based DSSC [15]. These natural dyes have the advantage of quick charge transmission and allow the use of high-viscosity electrolytes like ionized electrolytes.

This research sought to investigate the performance of naturally existing dyes as sources of biological sensitizers in the development of dye-sensitized solar cells (DSSCs). Also Carbon counter electrode was used instead of Platinum (Pt) to reduce the cost. The natural dyes were taken out from *Tradescantia pallida (Purplequeen), Jungle geranium* (Rongon), and *Catharanthus roseus* (Noyon tara). The DSSCs using natural dyes as sensitizer usually shown lower power conversion efficiency (PCE) than Ruthenium (N3) synthetic dye. Nevertheless natural dyes was used in this work because of toxicity and disposal challenging of the Ruthenium complex dye.

2. Methodology

2.1 Preparation of Working Electrode

As the substrate, fluorine-doped tin oxide (FTO) glass was chosen since it had a resistance of 8 ohms per square (Ω/□) (Solaronix). Combining ethyl alcohol and de-ionized water, the FTO glass was rigorously cleaned before being dried with an ultrasonic cleaner and N_2 gas [5]. The purified FTO was then coated with successive layers of nanostructured titanium dioxide (TiO_2) using a spin coater. To make the TiO_2 film, a dissolving consisting of TiO_2, 0.5g acetyl-acetone, 0.1 ml, 4 ml of a water-ethanol mixture, and 0.4 ml of triton X-114 was employed. Spinning at a speed of 3000 rpm for 5 minutes was required for the spin coating process. The completed film was then dried to 450 °C for an hour in a resistive furnace box after being left at the temperature of the room for nearly an hour [5]. By using an X-ray diffractometer (SHIMADZU XRD-6000) and the Cu-Ka line, grazing incident X-ray diffraction (GIXRD) analysis was used to determine the crystal structures of the TiO_2 thin films. The UV-VIS spectrophotometer was used to measure the optical characteristics of the TiO_2 films. A field-emission scanning electron microscope (FE-SEM) (JEOL, JSM-6700F) was used to examine the surface morphologies. **Figure 1** shows the process flowchart for preparing the TiO_2 working electrode.

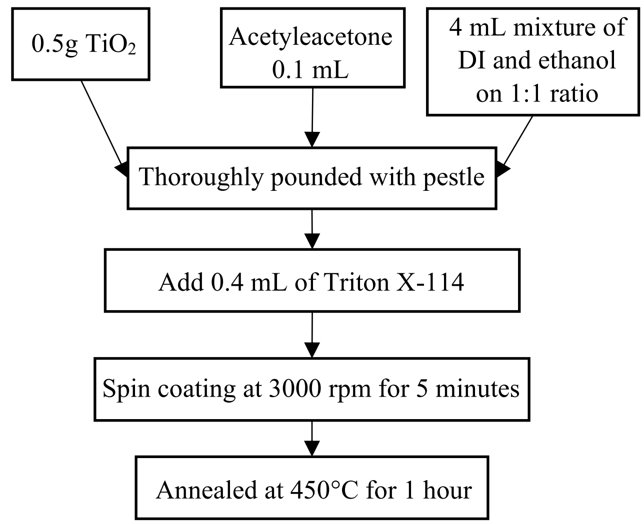


Figure 1. Process flowchart for preparing the TiO_2 working electrode

2.2 Preparation of Counter Electrode

A mixed mixture of carbon powder, carbon black, and carboxyl methyl cellulose weighing, respectively, 1 g, 0.2 g, and 0.32 g were used to make a carbon counter electrode using the doctor blade method [4]. Along with the aforementioned chemical components, 16 ml of de-ionised water and 8 ml of ethyl alcohol were added. The whole cap area of the carbon black was just 68 m^2/g, compared to the used carbon powder's overall surface area of 1100 m^2/g. Prior to being put on the conductive substrate of glass for FTO, each component was thoroughly mixed together. The created carbon sheet was then heated at 175 °C for an hour [6]. **Figure 2** shows the process flowchart for preparing the Carbon counter electrode.

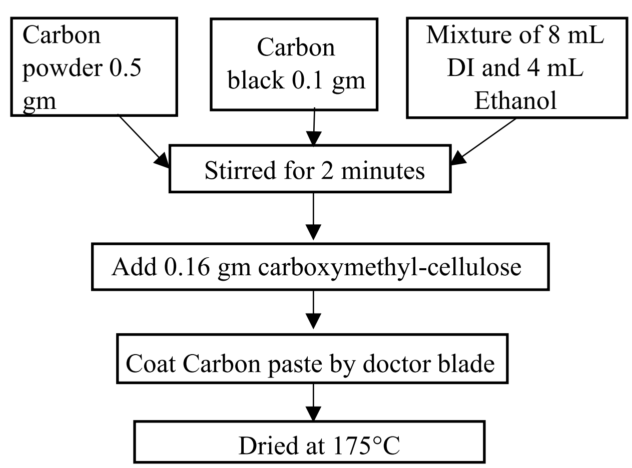


Figure 2. Process flowchart for preparing the carbon counter electrode

2.3 Preparation of Electrolyte

Electrolyte plays a very crucial role in DSSC. The functions of electrolytes in DSSC are electron transport, redox mediator, and dye molecule regeneration. An electrolyte was prepared by mixing KI and I_2 at molar concentrations of 0.5 M and 0.05 M, respectively, with ethylene glycol [5]. Also, 0.05M 4 tert-butylpyridine was added to enhance electron mobility. **Figure 3** shows the process flowchart for preparing electrolytes.

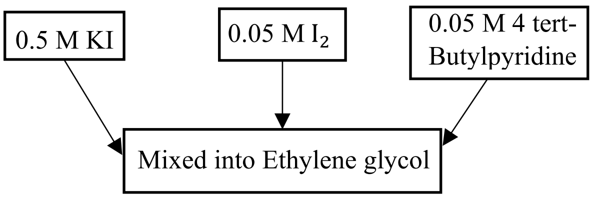


Figure 3. Electrolyte preparation process

2.4 Preparation of Natural Dyes

The natural dye from three different sources had been extracted from leaves and flowers using standard preparation procedure.

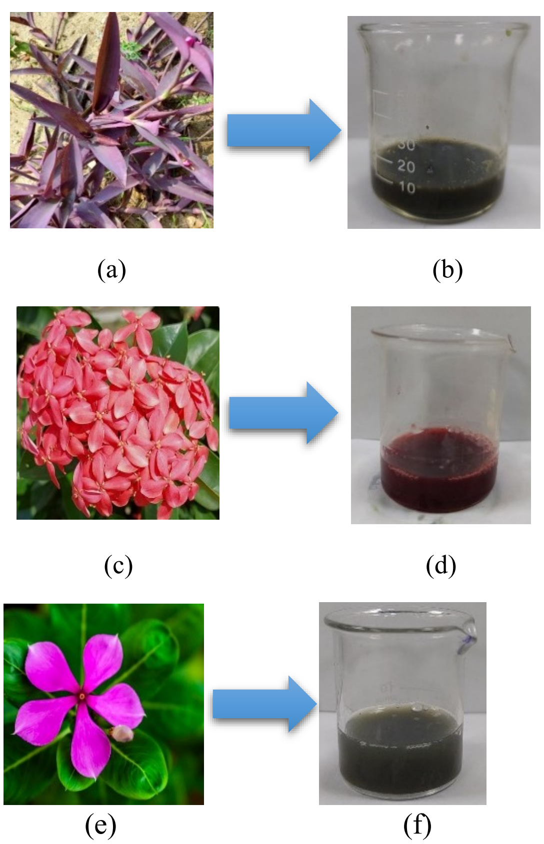


Figure 4. (a) Images of (a) *Tradescantia pallida* (Purple Queen) leaves, (b) extracted dye, (c) *Jungle Geranium* (Rongon) flower, (d) extracted dye, (e) *Catharanthus roseus* (Noyon tara), (f) extracted dye

Initially, distilled water was used to clean the leaves of the *Tradescantia pallida (*Purple queen*)* plant, and the flowers of *Jungle geranium (*Rongon*) as well as Catharanthus roseus (*Noyon Tara*)*. The leaves and flowers were then ground with a mortar and pestle into a paste. The dyes were then cordially extracted from the composition and kept in a beaker after being pressurized by hand while being wrapped in a cotton cloth. The dyes were applied as a photosensitizing agent after filtering. **Figure 4** shows the dye extraction process.

2.5 DSSC Integration

The TiO_2 porous films were first all dye-sensitized prior to being exposed to the dyes, all of the TiO_2 transparent films were dye-sensitized. The dye-sensitized TiO_2 photoelectrode (anode) was sandwiched between a carbon counter electrode (cathode), creating the DSSCs. The space across the two electrodes was filled with electrolytes [6]. To keep the solar cell from shorting out, an electrode spacer was placed between them. A 0.25 cm^2 active cell surface was present in the cell. A I-V curve tracker (Keithley-2400) was used to determine the solar cells current versus voltage (I-V) characteristics while it was operating under the impact of AM 1.5 sun irradiation.

3. Results and Discussion

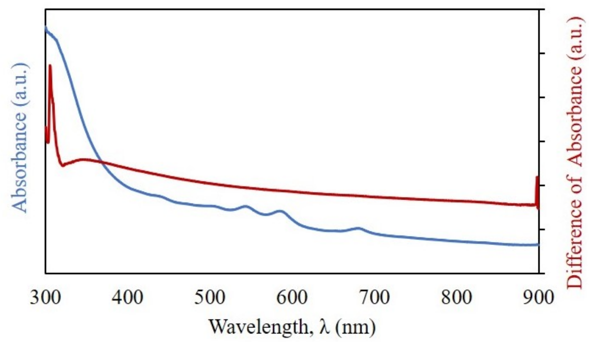
The optical absorption spectra of the Purple Queen, Rongon, and Noyon Tara dyes and TiO_2 film were measured using a UV-VIS spectrophotometer [6]. From **Figure 5**, it is observed that the dye absorbs the majority of photons in between 300nm to 700nm of wavelengths. In this region, many small peaks have observed between 500-700 nm, 450-550 nm, and 650-700 nm of wavelengths. These peaks indicate about moving the electrons into the dye's conduction band at higher level. The difference between the absorbing capacity of TiO_2 films sensitized with dye and those that are not was also examined using the absorbency of dye in "**Figure 5**". This comparison makes it obvious that the TiO_2 coating has absorbed the dye.

The tauc plot is a widely used technique for calculating the bandgap energy of a material using UV-VIS absorption spectroscopic data [6]. The formula for the equation is

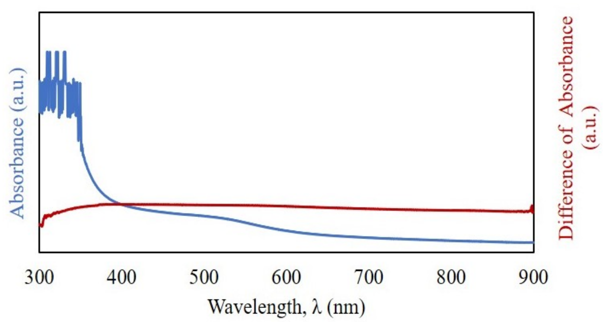
$$(\alpha h\upsilon) = A\,(h\upsilon - E_g)^n \quad \text{(i)}$$

Where, n=1/2 for the direct bandgap and n=2 for the indirect bandgap.

From the intercept of the linear component of the function of $(\alpha h\upsilon)^2$ versus the photon energy ($h\upsilon$), where $h\upsilon$ is the photon energy and α is the absorption coefficient, one may calculate the bandgap energy [11]. The bandgap energy for the Purple queen, Rongon and Noyon Tara dyes are respectively 3.25 eV, 3.2 eV and 3.08 eV which is shown in **Figure 6**.



(a)



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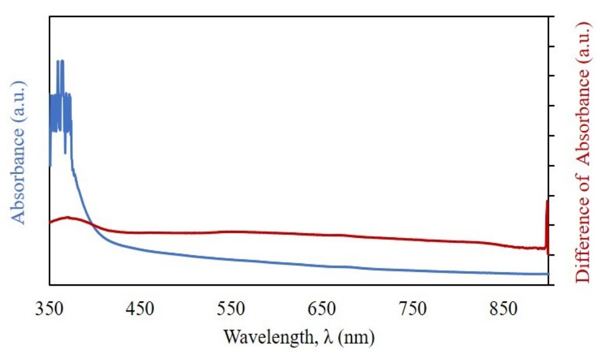


(c)

Figure 5. The spectrum of optical absorption of (a) *Tradescantia pallida* (Purple Queen) (b) *Jungle Geranium* (Rongon) (c) *Catharanthus roseus* (Noyontara) dye and variation in absorption of TiO_2 working electrode between with and without dye sensitization

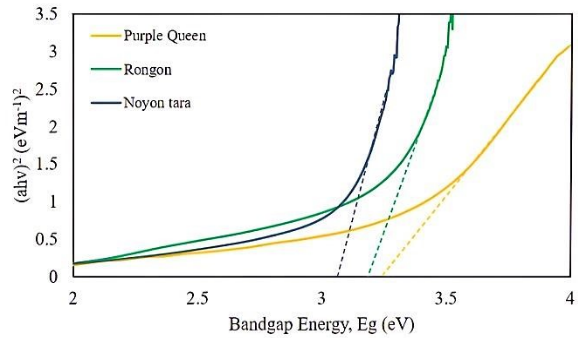
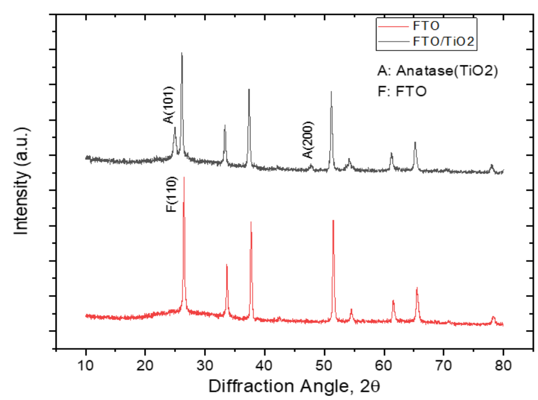


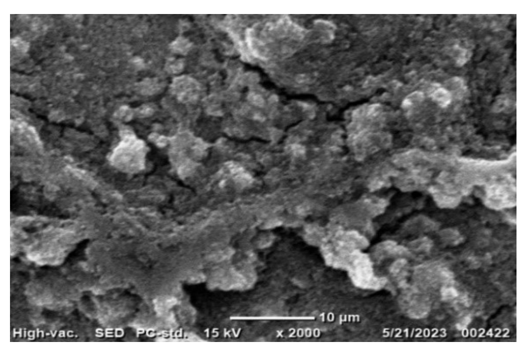
Figure 6. Bandgap energy plot for *Tradescantia pallida* (Purple Queen), *Jungle Geranium* (Rongon), *and Catharanthus roseus* (Noyontara) dye

The bandgap energy of *Tradescantia pallida* (Purple Queen) dye is 3.25 eV, for Jungle Geranium (Rongon) dye 3.2 eV, and for Catharanthus roseus (Noyontara) dye 3.08 eV.

The X-ray diffraction patterns for the TiO_2 electrode with an incidence angle, $\alpha = 0.5°$ are indicated in "**Figure 7 (a)**". These spikes demonstrated the TiO_2 film's polycrystalline nature.



(a)



(b)

Figure 7. Characterization of TiO_2 film X-ray diffraction pattern in (a), with an inset of an EDS analysis FE-SEM picture in (b).

With the highest peak, A(101), exhibit at an angle of 25.30°, the majority of spikes were linked with the anatase phase. A(200) was traced at a 2θ value of 46.07°.

The Debye-Scherrer formula,

$$D = \frac{0.94\lambda}{\beta\cos\theta} \quad \text{(ii)}$$

The crystal size (D) of the TiO_2 film was discerned using the X-ray dispersion wavelength (λ) of Cu Kα (0.154 nm) and the diffraction angle (θ), with the full width at half maximum (FWHM) denoted by β [6].

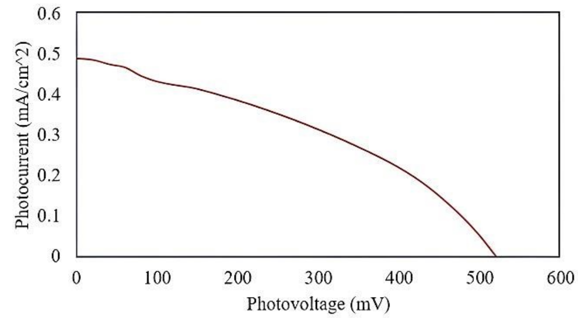
The anatase peak A(101) was used for the finding of crystal dimensions, which is 14.9 nm. Using the FE-SEM technics, the surface properties of the crystalline TiO_2 coating was investigated and are shown in " **Figure 7 (b)**". The FE-SEM image depicture the porous shape of the TiO_2 layer with distinct grain clusters.

The voltage versus current density plot of the three dye-sensitized solar cells have been shown in **Figure 8**. The cell performance can be calculated from the following equations [6],

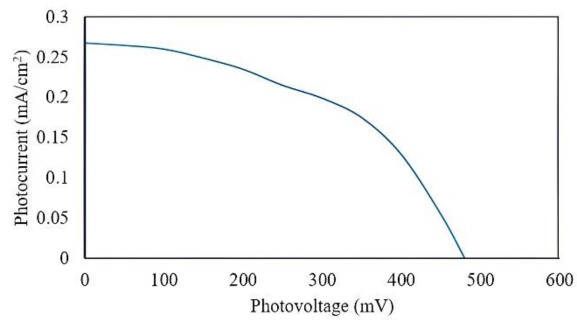
$$\eta = (Voc*Jsc*FF)/Pin \quad \text{(iii)}$$

$$\text{where, } FF = (Vmax*Jmax)/(Voc*Jsc) \quad \text{(iv)}$$

Jsc, Voc, and FF imply the short circuit current density, open circuit voltage, and fill factor respectively. Pin (100 mW/cm^2) represents the magnitude of incident light illumination. In case of *Tradescantia pallida* (Purple Queen) based DSSC, Voc of 520 mV, Jsc of 0.531 mA/cm^2, FF of 34.4%, and cell efficiency of 0.095% were found. in case of *Tradescantia pallida* (Purple Queen) based DSSC. In case of *Catharanthus roseus* (Noyontara) based DSSC, Voc of 480 mV, Jsc of 0.267 mA/cm^2, FF of 46.74%, and cell efficiency of 0.0611% were observed. Finally, in case of *Jungle Geranium* (Rongon) based DSSC, Voc of 345 mV, Jsc of 0.31 mA/cm^2, FF of 49.37%, and cell efficiency of 0.0524% were investigated. The maximum power conversion efficiency (PCE) was observed 0.095% in case of *Tradescantia pallida* (Purple Queen) based DSSC. Though the *Jungle Geranium* (Rongon) based DSSC showed highest fill factor (FF) , still lower efficiency was observed due to lower level of open circuit voltage and short circuit current. All these open circuit voltages and short circuit currents have been measured using Keithley-2400 I-V curve tracker module under the 1.5 AM illumination of solar simulator. The solar cells performance metrics are displayed in **Table 1**.

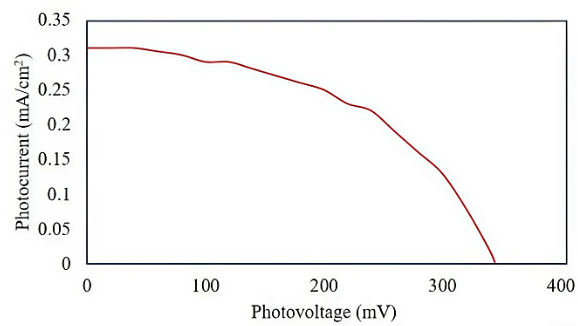


(a)



(b)

Figure 8. Photovoltage vs (c) photocurrent density



(J-V) Characteristic Curve of (a) *Tradescantia pallida* (Purple Queen) (b) *Jungle Geranium* (Rongon) (c) *Catharanthus roseus* (Noyontara) based dye-sensitized solar cells (DSSCs)

4. Conclusion

The incorporation of natural pigments in DSSCs has unveiled substantial potential for enhancing solar energy harness. This investigation has expounded upon the intricate nexus between the pigment derived from organic sources, such as *Tradescantia pallida* (Purple Queen) leaves, *Catharanthus roseus* (Noyontara) flowers, and *Jungle Geranium* (Rongon) flowers and the photoelectrochemical performance within the solar cell. The fusion of DSSC, catalysed by the aforenamed pigment, has exhibited notable photovoltaic characteristics. The observed conversion efficiency of 0.095% for *Purple Queen* leaves-based DSSC, 0.0611%

Catharanthus roseus (Noyontara) flowers-based DSSC, and 0.0528% for *Jungle Geranium* (Rongon**)** flowers-based DSSC validates the feasibility of employing natural pigments as sensitizing agents in DSSCs. Higher efficiency and stability can be obtained by using solid state electrolyte and Platinum counter electrode. This materials couldn't be used in this work due to unavailability of these materials in the lab.

Table 1. Performance analogy of DSSCs

Sensitizer	Voc (mV)	Jsc (mA/cm^2)	FF (%)	η (%)	Reff.
Tradescantia pallida	520	0.531	34.4	0.095	This work
Catharanthus roseus	480	0.267	46.74	0.0611	This work
Jungle Geranium	345	0.31	49.37	0.0528	This work
Basella Alba	383	0.222	37.6	0.032	[5]
Oleander	330	0.55	36	0.07	[7]
Pink Frangipani	310	0.55	30	0.05	[7]
Mulberry	520	0.181	54.19	0.051	[8]
Red cabbage	510	0.21	46.61	0.06	[9]
Strobilanthes cusia	306.35	0.0155	46.2	0.0385	[10]
Green cabbage	456	0.223	44.19	0.095	[15]

5. Acknowledgement

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