



## سنتر نانوذرات $\text{TiO}_2$ با روش سل-ژل برای ساخت سلول‌های خورشیدی حساس شده با رنگدانه طبیعی میوه جمبو

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**چکیده-** در این مطالعه، نانوذرات  $\text{TiO}_2$  به منظور دستیابی به بلورینگی بالا و اندازه کوچک ذرات، به روش سل-ژل سنتز شده و سپس مشخصه‌های نانوذره‌های تهیه شده به وسیله پراش پرتو ایکس (XRD) و طیف سنج مادون قرمز (FT-IR) ارزیابی شدند. سلول خورشیدی رنگدانه‌ای با استفاده از لایه نشانی خمیر نانوذرات  $\text{TiO}_2$  بر روی زیرلایه شیشه‌ای FTO (اکسید قلع آلیبده شده با فلورین) تحت عنوان فیلم اکسید نیمه رسانا ساخته شد. همچنین در این تحقیق، برای کاهش هزینه‌ها، از رنگدانه طبیعی استخراج شده از میوه جمبو استفاده شده است. پارامترهای فتوولتائیک تحت نور خورشید شبیه سازی شده با شدت تابش  $100 \text{ mW/cm}^2$  و تابش استاندارد ( $\text{AM}1.5$ ) اندازه گیری شد. نتایج نشان داد بازده تبدیل انرژی  $1.36\%$  و سایر مشخصه های فتوولتائیک شامل جریان مدار کوتاه، ولتاژ مدار باز و فاکتور پر شدن به ترتیب  $5.88 \text{ mA/cm}^2$ ،  $0.6235 \text{ V}$  و  $0.3706$  می‌باشند.

**کلید واژه-** رنگدانه طبیعی، روش سل-ژل، سلول‌های خورشیدی رنگدانه‌ای، فتوآند، نانوذرات  $\text{TiO}_2$

### Synthesis of $\text{TiO}_2$ nanoparticles using the sol-gel method to fabricate dye-sensitized solar cells with natural Syzygium Cumini fruit dye

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**Abstract** In this study,  $\text{TiO}_2$  nanoparticles were synthesized using the sol-gel method to obtain a highly crystalline and small particle size of  $\text{TiO}_2$ ; the produced nanoparticles were characterized by X-ray diffraction (XRD) and Fourier transform infrared spectroscopy (FT-IR). Dye-sensitized solar cell (DSSC) was fabricated with a layer of  $\text{TiO}_2$  nanoparticles paste deposited on FTO (fluorine-doped tin oxide) glass substrate as a semiconductor oxide film. Also in this study, extracted natural dye from Syzygium Cumini fruit was used for decrease the cost. The photovoltaic parameters were measured using the solar simulator under an incident light intensity of  $100 \text{ mW/cm}^2$  and air mass ( $\text{AM}1.5$ ). The results show power conversion efficiency ( $\eta$ ) of  $1.36\%$  and other photovoltaic parameters include short circuit current density ( $J_{sc}$ ), open-circuit voltage ( $V_{oc}$ ) and fill factor ( $FF$ ) are  $5.88 \text{ mA/cm}^2$ ,  $0.6235 \text{ V}$  and  $0.3706$  respectively.

**Keywords:** Dye-sensitized solar cells, Natural dye; Photoanode, Sol-gel method;  $\text{TiO}_2$  nanoparticles

## 1. Introduction

Dye-sensitized solar cells (DSSCs) have attracted considerable attention due to their low cost and easy fabrication with relatively high photo-conversion efficiency. O'Regan and Grätzel fabricated TiO<sub>2</sub> based DSSC for the first time in 1991 [1]. A typical DSSC is made up of semiconductor oxide film for attaching dye molecules, a counter electrode with deposited layer of platinum and an electrolyte solution. Sun light is absorbed by dye molecules and then electrons are injected to the conduction band of semiconductor oxide. Meanwhile oxidized dye molecules are regenerated by electrolyte solution also electrolyte ions regenerated through counter electrode. Semiconductor oxide film is the heart of DSSCs and the most studied materials are TiO<sub>2</sub>, ZnO and SnO<sub>2</sub> which TiO<sub>2</sub> has announced as the best one due to its unique properties and various advantages such as photochemical stability, high band gap (~3.2 eV), excellent optical transparency capability, availability, and non-toxicity. TiO<sub>2</sub> exists in three main phases, namely, rutile, anatase and brookite. However rutile phase is more thermal stable, anatase phase is the first choice for DSSCs applications due to its higher band gap energy [2, 3]. Various methods such as sol-gel, hydrothermal, solvothermal and etc. which sol-gel is one of the most used methods due to highly crystalline and small size of synthesized nanoparticles [4-6]. The sol-gel is a simple, fast, and cost-effective method, which has received much attention due to providing controlled grain size as well as particle morphology, achieving superior purity, compositional homogeneity, low processing temperature, and production with simple equipment. In this work, natural dyes were used to reduce costs and TiO<sub>2</sub> nanoparticles were synthesized using sol-gel method. Then, DSSC was fabricated based on synthesized TiO<sub>2</sub> and photovoltaic performance was evaluated under AM 1.5 G by measuring current-voltage curves and calculating  $\eta$ ,  $V_{oc}$ ,  $J_{sc}$ , and  $FF$ .

## 2. Experimental

### 2.1. Materials

The Titanium (IV) isopropoxide (Ti[OCH(CH<sub>3</sub>)<sub>2</sub>]<sub>4</sub>), ethanol (C<sub>2</sub>H<sub>5</sub>OH), distilled water, nitric acid (HNO<sub>3</sub>), polyethylene glycol (C<sub>n</sub>H<sub>2n+2</sub>O<sub>n+1</sub>), acetonitrile (C<sub>2</sub>H<sub>3</sub>N), potassium iodide (KIO<sub>3</sub>) and iodine (I<sub>2</sub>), ethylene glycol (C<sub>2</sub>H<sub>4</sub>O<sub>2</sub>), platinum (Pt). Syzygium Cumini fruit and fluorine-doped tin oxide (FTO) conductive glass (sheet resistance 10 Ω/sq).

### 2.2. Preparation of TiO<sub>2</sub> nanoparticles paste

In this method, TiO<sub>2</sub> nanoparticles were synthesized in anatase phase. First, titanium (IV) isopropoxide was added to ethanol under stirring and after few minutes, distilled water was added. Then adding few drops of nitric acid was done to control the pH of prepared solution. To form sol, solution must be stirred vigorously for 30 min, then aging for 24 hrs to obtain gel. For preparing TiO<sub>2</sub> nanoparticles from gel, it must be dried at 120 °C then, sintered at 450 °C to get white powder [4]. The procedure of TiO<sub>2</sub> synthesis is shown in Fig. 1. Finally, TiO<sub>2</sub> powder and polyethylene glycol were mixed into the mortar until uniform paste was obtained [7].

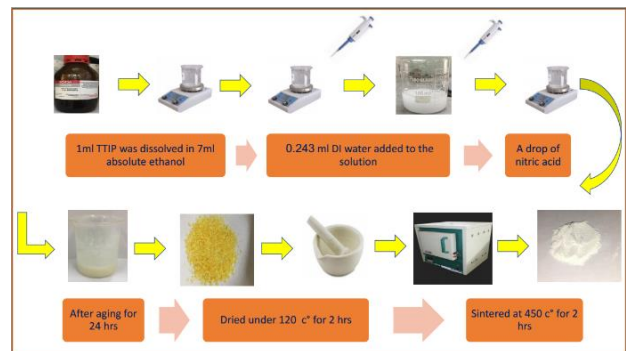


Fig. 1: The schematic of the synthesis route for TiO<sub>2</sub> via sol-gel method

### 2.3. Preparation of natural dye and electrolyte

It was extracted from fresh Syzygium Cumini fruit and ethanol was used as solvent [8, 9]. For preparation of electrolyte solution, first, 10 ml acetonitrile was added to 30 ml ethylene glycol under stirring. Subsequently, 1.370 g potassium iodide and 0.337 g iodine was added

respectively. Prepared electrolyte was stirred until homogenous solution appeared [9].

#### 2.4. Fabrication of DSSC

First, FTO glass substrate ultrasonically cleaned in deionized water, hydrochloric acid, acetone and ethanol respectively and dried at 70 °C. Afterwards, the prepared TiO<sub>2</sub> paste was coated on FTO glass, to make photoanode, by doctor blade method and after a few minutes it was heated at 120 °C then, calcinated at 450 °C. After cooling, the photoanode was immersed in dye solution and kept in darkness for 24 hrs. For the counter electrode, a thin layer of platinum was deposited on another FTO glass substrate. Finally, photoanode and counter electrode were combined together and sealed using surlyn sheet and electrolyte was injected between them. The active area of the electrode was 0.20 cm<sup>2</sup>. The photograph of the fabricated DSSC is given in Fig. 2.

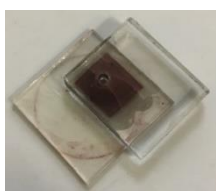


Fig. 2: The fabricated DSSC

### 3. Results and Discussion

Figure 3 shows the UV-vis absorption spectra in the range of 300–800 nm, which is shown a peak in 380 nm. The absorption spectra become sharper and the absorbance increases. The FT-IR spectroscopy of the sample were studied which were synthesized via sol-gel method in the range of 4000–400 cm<sup>-1</sup> and shown in Fig. 4. In this curve, peaks at 425 cm<sup>-1</sup> and 732 cm<sup>-1</sup> are for O–Ti–O bonding in anatase morphology. The bands centered at 2957 cm<sup>-1</sup> and 3182 cm<sup>-1</sup> are the characteristic of surface-adsorbed water and hydroxyl groups. Existing fine peaks also relate to the residual components of organic matter and reactions between water and carbon dioxide, while

the last peak is attributed to the TiO<sub>2</sub>. As well, XRD pattern was done to determine crystal structure of the prepared TiO<sub>2</sub> powder. The XRD peaks in the range of 2θ from 20°–90°, where the peaks in 20.418°, 37.208°, 38.143°, 48.226°, 54.344°, 55.292°, 63.080°, 70.087°, 75.070°, and 83.114° can be attributed to the 101, 103, 112, 200, 100, 211, 204, 220, 210, and 222 crystalline structures of anatase. This pattern is represented in Fig. 5.

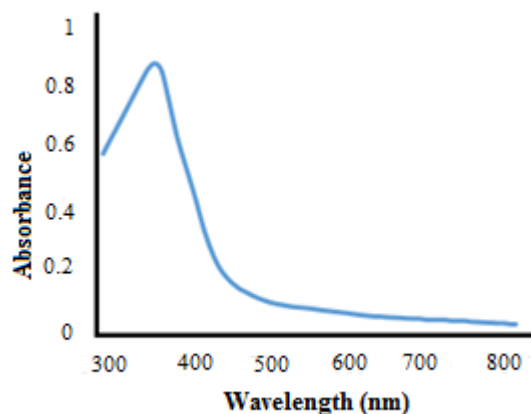


Fig. 3: The UV-visible of the fabricated DSSC

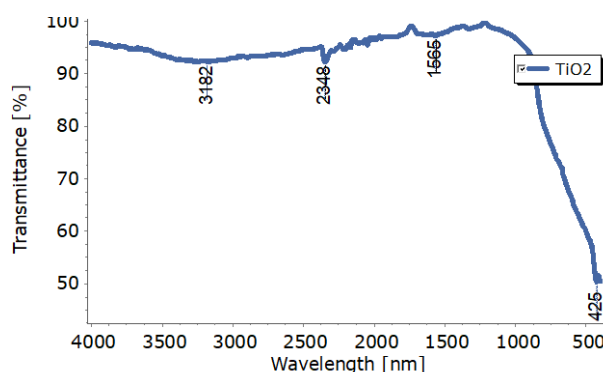


Fig. 4: The FTIR of the fabricated TiO<sub>2</sub> nanoparticles

The current density–voltage (J–V) characteristics is represented in fig. 6 and photovoltaic parameters of fabricated DSSC are calculated from this curve with incident light intensity of 100 mW/cm<sup>2</sup> and AM 1.5. The efficiency of fabricated cell is 1.26% whereas calculated photovoltaic parameters such as short circuit current density (*J<sub>sc</sub>*), open circuit voltage (*V<sub>oc</sub>*) and fill factor (*FF*) are 0.88 mA/cm<sup>2</sup>, 0.122 V and 0.376, respectively and with good agreement with pervious works [8, 10]. The results approved the existence of free carriers by light

photons absorption. It is expected to enhance the efficiency of dye adsorption by increasing the grain boundaries of the produced TiO<sub>2</sub> using sol-gel method.

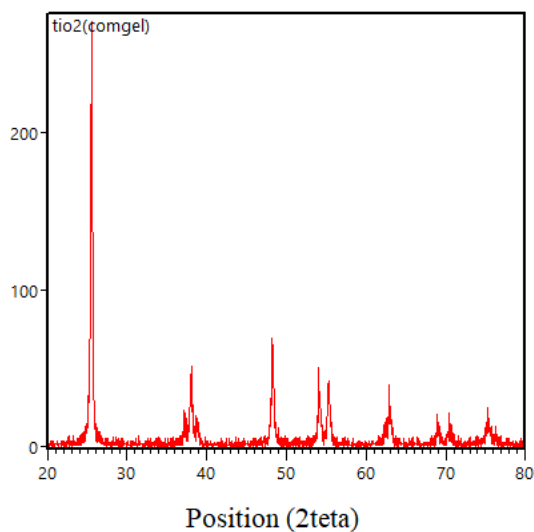


Fig. 5: Powder XRD pattern of the TiO<sub>2</sub> nanoparticles

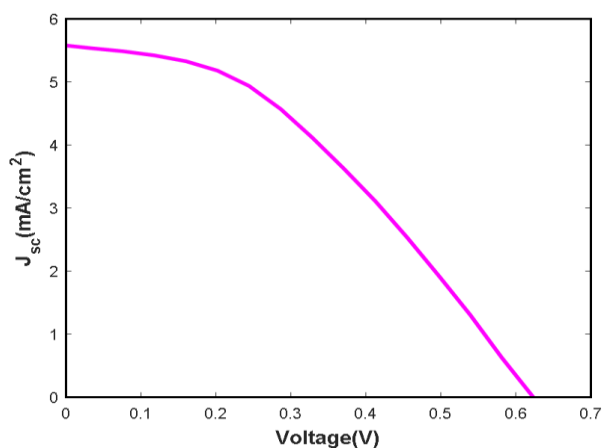


Fig. 6: current density–voltage curve of the fabricated DSSC

#### 4. Conclusion

TiO<sub>2</sub> nanoparticles have been successfully synthesized using sol-gel method. The synthesized TiO<sub>2</sub> nanoparticles were characterized using XRD, and FTIR techniques. The produced nanoparticles were used as a part of photoanode in the DSSCs. The pastes were prepared with simple method and used in the photoanode of the fabricated DSSCs. The crystallography of the pastes, using X-ray illustrated the existence of TiO<sub>2</sub> in the anatase phase in all samples. The fabricated TiO<sub>2</sub>-based DSSCs demonstrated a light to the electricity conversion

efficiency of 1.13% with a fill factor of 37.6%, open-circuit voltage of 0.65 V, and short-circuit current of 3.5 mA/cm<sup>2</sup>.

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