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# سنتز نانوذرات $TiO_2$ با روش سل-ژل برای ساخت سلول‌های خورشیدی حساس شده با رنگدانه طبیعی میوه جمبو

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**چکیده**- در این مطالعه، نانوذرات $TiO_2$ به منظور دستیابی به بلورینگی بالا و اندازه کوچک ذرات، به روش سل-ژل سنتز شده و سپس مشخصه‌های نانوذره‌های تهیه شده به وسیله‌ی پراش پرتو ایکس (XRD) و طیف سنج مادون قرمز (FT-IR) ارزیابی شدند. سلول خورشیدی رنگدانه‌ای با استفاده از لایه نشانی خمیر نانوذرات $TiO_2$ بر روی زیرلایه شیشه‌ای FTO (اکسید قلع آلاییده شده با فلوئور) 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 ۱۰۰ $mW/cm^2$ و تابش استاندارد (AM۱،٥) اندازه گیری شد. نتایج نشان داد بازده تبدیل انرژی ۱،۳۶٪ و سایر مشخصه های فتوولتایک شامل جریان مدار کوتاه، ولتاژ مدار باز و فاکتور پر شدن به ترتیب ٥،۸۸ $mA/cm^2$ ، ۰،۶۲۳٥ V و۰،۳۷۰۶ می‌باشند.

**کلید واژه-** رنگدانه طبیعی، روش سل-ژل، سلول‌های خورشیدی رنگدانه‌ای، فتوآند، نانوذرات $TiO_2$

# Synthesis of $TiO_2$ 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, $TiO_2$ nanoparticles were synthesized using the sol-gel method to obtain a highly crystalline and small particle size of $TiO_2$; 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 $TiO_2$ 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 ۱۰۰ $mW/cm^2$ and air mass (AM۱،٥). The results show power conversion efficiency (η) of ۱،۳٦٪ and other photovoltaic parameters include short circuit current density ($J_{SC}$), open-circuit voltage ($V_{OC}$) and fill factor ($FF$) are ٥،۸۸ $mA/cm^2$, ۰،٦۲۳٥ V and ۰،۳۷۰٦ respectively.

Keywords: Dye-sensitized solar cells, Natural dye; Photoanode, Sol-gel method; $TiO_2$ nanoparticles

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## ۱. 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 $TiO_2$ based DSSC for the first time in ۱۹۹۱ [۱]. 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 $TiO_2$, ZnO and $SnO_2$ which $TiO_2$ has announced as the best one due to its unique properties and various advantages such as photochemical stability, high band gap (~۳,۲ □□), □□□□ □□□□□□ □□□□□□□□ capability, availability, and non-toxicity. $TiO_2$ 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 [۲, ۳]. □□□□□□□□□ □□□□□□□ □□□ □□□□□□□ □□□ □□□□□□□□□ nanocrystalline $TiO_2$ 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 [٤-٦]. 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 $TiO_2$ nanoparticles were synthesized using sol-gel method. Then, DSSC was fabricated based on synthesized $TiO_2$ and photovoltaic performance was evaluated under AM ۱,٥ G by measuring current-voltage curves and calculating $\eta$, $V_{oc}$, $J_{sc}$, and $FF$.

## ۲. Experimental

### ۲,۱. Materials

The Titanium (IV) isopropoxide ($Ti[OCH(CH_3)_2]_4$), ethanol ($C_2H_5OH$), distilled water, nitric acid ($HNO_3$), polyethylene glycol ($C_{2n}H_{4n+2}O_{n+1}$), acetonitrile ($C_2H_3N$), potassium iodide ($KIO_3$) and iodine ($I_2$), ethylene glycol ($C_2H_6O_2$), platinum (Pt). Syzygium Cumini fruit and fluorine-doped tin oxide (FTO) conductive glass (sheet resistance ۱٥Ω/sq).

### ۲,۲. Preparation of $TiO_2$ nanoparticles paste

In this method, $TiO_2$ 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 ۳۰ min, then aging for ۲٤ hrs to obtain gel. For preparing $TiO_2$ nanoparticles from gel, it must be dried at ۱۲۰ ºC then, sintered at ٤٥۰ ºC to get white powder [٤]. The procedure of $TiO_2$ synthesis is shown in Fig.۱. Finally, $TiO_2$ powder and polyethylene glycol were mixed into the mortar until uniform paste was obtained [۷].

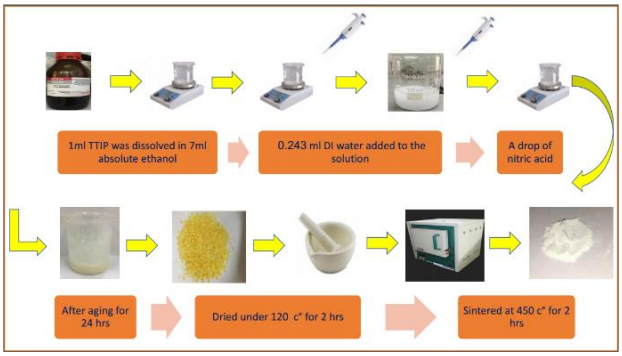



Fig. ۱: The schematic of the synthesis route for $TiO_2$ via sol-gel method

### ۲,۳. Preparation of natural dye and electrolyte

It was extracted from fresh Syzygium Cumini fruit and ethanol was used as solvent [۸,۱۰]. For preparation of electrolyte solution, first, ۱۰ ml acetonitrile was added to ۲,٥ ml ethylene glycol under stirring. Subsequently, ۱,۰۳۷٥ g potassium iodide and ۰,۳۳۰۷ g iodine was added

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respectively. Prepared electrolyte was stirred until homogenous solution appeared [۹].

### ۲,۴. Fabrication of DSSC

First, FTO glass substrate ultrasonically cleaned in deionized water, hydrochloric acid, acetone and ethanol respectively and dried at ۷۰ ºC. Afterwards, the prepared $TiO_2$ paste was coated on FTO glass, to make photoanode, by doctor blade method and after a few minutes it was heated at ۱۲۰ ºC then, calcinated at ٤٥۰ºC. After cooling, the photoanode was immersed in dye solution and kept in darkness for ۲٤ 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 ۰,۲٥ $cm^2$. The photograph of the fabricated DSSC is given in Fig.۲.

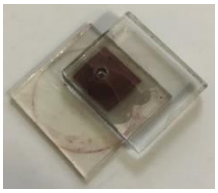


Fig. ۲: The fabricated DSSC

## ۳. Results and Discussion

Figure. ۳ shows the UV-vis absorption spectra in the range of ۳۰۰-۸۰۰ nm, which is shown a peak in ۳۸۰ □□.□□ □□□ □□□□ □□ □□□□□□□□□□ □□□□□□□□□□, □□□□□ 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 ٤۰۰–٤۰۰۰ $cm^{-1}$.and shown in Fig.٤. In this curve, peaks at ٤۲٥ $cm^{-1}$ and ۷۳۲ $cm^{-1}$ are for O–Ti–O bonding in anatase morphology. The bands centered at ۲۰٤۷ $cm^{-1}$ and ۳۱۸۲ $cm^{-1}$ 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 $TiO_2$. As well, XRD pattern was done to determine crystal structure of the prepared $TiO_2$ powder. The XRD peaks in the range of $2\theta$ from ۲۰°-۹۰°, where the peaks in ۲٥,٤۱۸°, ۳۷,۲۰۸°, ۳۸,۱٤۳°, ٤۸,۲۲٦°, ٥٤,۳٤٤°, ٥٥,۲۹۲°, ٦۳,۰۸٥°, ۷۰,٥۸٦°, ۷٥,٥۷٥°, and ۸۳,۱۱٤° can be attributed to the ۱۰۱, ۱۰۳, ۱۱۲, ۲۰۰, ۱۰٥, ۲۱۱, ۲۰٤, ۲۲۰, ۲۱٥, and ۲۲٤ crystalline structures of anatase. This pattern is represented in Fig.٥.

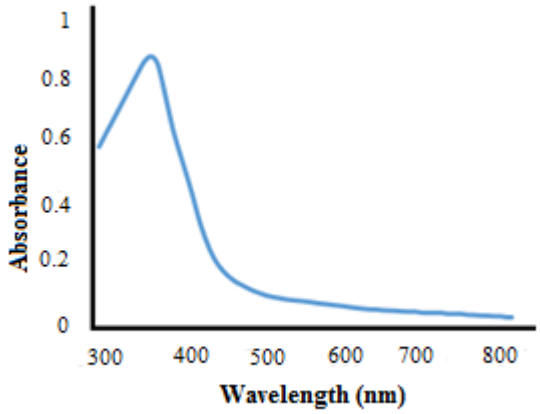



Fig. ۳: The UV-visible of the fabricated DSSC

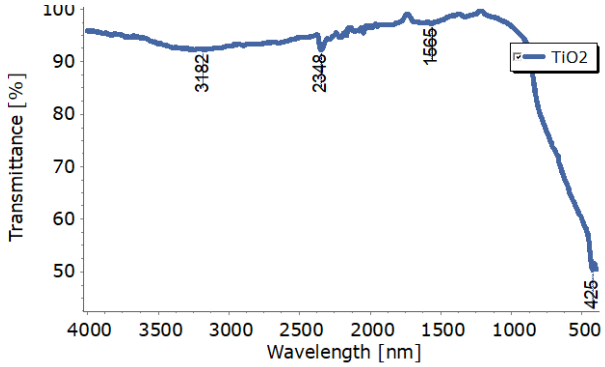



Fig. ٤: The FTIR of the fabricated $TiO_2$ nanoparticles

The current density–voltage (J–V) characteristics is represented in fig. ٦ and photovoltaic parameters of fabricated DSSC are calculated from this curve with incident light intensity of ۱۰۰ mW/$cm^2$ and AM ۱,٥. □□□ □□□□□□□□ □□□□□□□ □□□□□ □□□□□ □□□□□□□□□□□ efficiency of fabricated cell is ۱,۳٦٪ whereas calculated photovoltaic parameters such as short circuit current density ($J_{SC}$), open circuit voltage ($V_{OC}$) and fill factor ($FF$) are ٥,۸۸ mA/$cm^2$, ۰,٦۲۳٥ V and ۰,۳۷۰٦, respectively and with good agreement with pervious works [۸,۱۰]. The results approved the existence of free carriers by light

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photons absorption. It is expected to enhance the efficiency of dye adsorption by increasing the grain boundaries of the produced $TiO_2$ using sol-gel method.

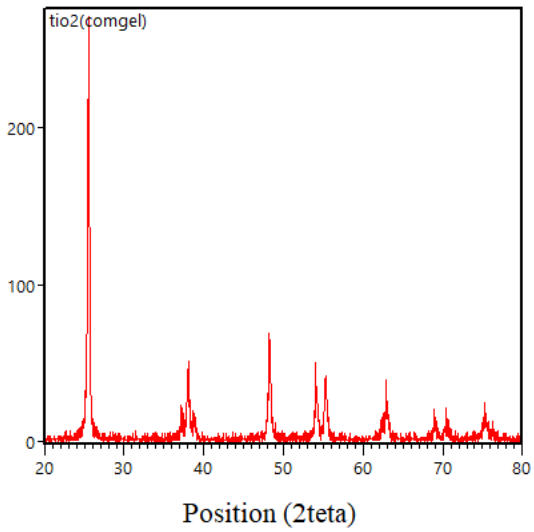



Fig. ٥: Powder XRD pattern of the $TiO_2$ nanoparticles

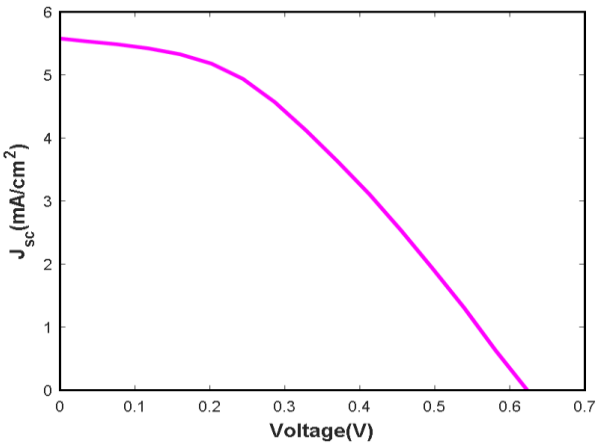



Fig. ٦: current density–voltage curve of the fabricated DSSC

## ٤. Conclusion

$TiO_2$ nanoparticles have been successfully synthesized using sol-gel method. The synthesized $TiO_2$ 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 $TiO_2$ in the anatase phase in all samples. The fabricated $TiO_2$-based DSSCs demonstrated a light to the electricity conversion efficiency of ١,١٣٪ with a fill factor of ٣٧,٥٪, open-circuit voltage of ٠,٦٢ V, and short-circuit current of ٣,٧ mA/cm$^2$.

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