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Preparation of Polyaniline/TiO_2 Photovoltaic Solar Cell

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ABSTRACT

Polyaniline (PANI) and Kronos C doped Titanium dioxide (TiO_2) was fabricated as PANI/TiO_2 solar cell and reported on its simple photovoltaic performance detected by using voltameter and tested for stability for 12 months. The PANI and TiO_2 were coated onto different indium tin oxide (ITO) glass plates by using a drop-casting method and sandwich attached for simple solar cell preparation. PT4 sample was the optimum solar cell with 0.2: 0.2 g of PANI: TiO_2 ratio under 2 cm^2 surface area based on the highest voltage produces from 100 mW cm^{-1} of light intensity metal highlight lamp. No voltage was detected for PT7 where the single layer TiO_2 coated without PANI attached for solar cell system using same fabrication. It shows that PANI has a significant role in functionalizing the photovoltaic system. The C-N stretching of aromatic amine and C-N stretching for the benzenoid at peaks of 1222 and 1166 cm^{-1} respectively in FTIR spectra has confirmed the PANI structure supported by an XRD pattern. TP4 has the highest photovoltaic performance compared to other types of TiO_2 based on the electron lifetime (τe) and the voltage produced was sustained up to 12 months.

Keywords: *Polyaniline, titanium dioxide, photovoltaic, solar cell*

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INTRODUCTION

Solar energy is sort of feasible and essential energy resource for all human beings, animals, and plants on earth. The renewable energy sources, ie; wind, hydro, geothermal, solar heating, solar photovoltaic, tidal, biomass and wave. Solar cell is well recognized to have significance potential as future energy source of demand [1,2]. Solar energy produces electric power without carbon emission, thus contributing to cleaner air and fewer greenhouse gases [3]. To deal with environmental problem and energy shortage, solar photovoltaic (PV) technology has been developed which is directly generate electricity from solar energy. In the advancement of new materials and structures, a massive attention has been given to modify inorganic oxide and the organic conjugated polymer [4].

Apart from commercially available silicone PV solar cell, titanium dioxide (TiO_2) nanoparticles also have demonstrated potential for solar cells implementation. In PV cell application, TiO_2 nanomaterial is a suitable candidate due to its wide stability of chemical and optical, non-toxicity, low cost, resistance to corrosion and ease of synthesis [5,6]. Dye-sensitized solar cell (DSSC) discovered by Grätzel [7] has emerged the potential of TiO_2 as important element in early stage of PV. In the contrary, the photoactivity of TiO_2 is restricted to ultraviolet (UV) region because of the wide band gap energy [8-14]. To overcome the band gap issue, researchers made attempts to broaden the optical response of TiO_2 to visible light by surface modification, doping with metal or non-metal, dye or semiconductor sensitization [15-17].

TiO_2 also has been diversely used in white paint pigment, health and beauty products [18]. Several studies have reported that the TiO_2 modification with carbon (C) named as C doped TiO_2 has gain a significant of interest. C doped TiO_2 reduces the bandgap energy making it active under wide optical respond [19-21]. However, the preparation of C doped TiO_2 only can be prepared by titania precursor under sol gel method, and this makes the process of preparing C doped TiO_2 is complicated. Recently, commercial C doped TiO_2 can be easily obtained and KRONOS VLP7000 is one of the most stable C doped TiO_2 available at the market. Reactive red 4 (RR4) dye as a TiO_2 sensitizer has also being studied by a few researchers [22-24] where the dye can act as an electron ejector to enhance photocatalytic activity in degrading other dyes [22-24].

Polyaniline (PANI) is one of the most versatile heterogeneous-conjugated polymers due to the capability to substitute metals and non-metal for TiO_2 photocatalytic efficiency [25]. PANI also has potential applications in alcohol dehydration [26-27] adsorbents, supercapacitors [28], sensors [29], pervaporation applications [30-31] and multiwalled carbon nanotubes (MWCNTs) coating [32]. This conducting polymer is studied for different redox states for electrochemical energy storage system mostly because of the long cycle stability and good theoretical specific capacities [33]. Besides its good electrical conductivity, PANI can also be simply coated on conductive glass by sol-gel and other physical methods such as dip coating, brush coating and drop

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casting techniques [34]. PANI is also useful due to its reversible redox behaviors which commonly used for supercapacitor, gas sensor, pH sensor and fuel cell application [35].

Therefore, PANI is a good choice for the modification of TiO_2 for photocatalytic applications. It represents as holes scavenger and improves photocatalytic activity when electrons are excited into the photocatalyst conduction band under visible light irradiation [36]. PANI has also potential to be used as a counter electrode in PV since it can attract as a hole acceptor [37]. Studies have found that polymer that contains a dopant can significantly change the electrochemical properties of the prepared polymer composite [38]. Thus, a less practical liquid electrolyte can be replaced by solid PANI.

PANI on TiO_2 as a solar cell was prepared by other researchers and reported it has a significant effect in photovoltaic [39]. For instance, Nemade et al. has reported that the efficiency associated with 15 wt% PANI loaded graphene composite, improved further by addition of TiO_2 nanoparticles [40]. While Bahramian and Vashaee reported that the combination of PANI and TiO_2 enhanced the light absorption with 8.22% efficiency of the fabricated bifacial dye-sensitized solar cel (DSSC) corresponding to the both-side irradiation [41]. Furthermore, a study by Zhou et al. revealed that enhanced photocurrent and good stability are the result of synergic effect between PANI and TiO_2 in which PANI may promotes the charge separation efficiently [42].

However, there is still no focus on the stability study of PANI/TiO_2 of solar cell being carried out. The aim of this study is to evaluate the voltage sustainability produced from the prepared PANI/TiO_2 sandwiched system, characterization, and optimization evaluations of the prepared PANI/TiO_2 with different types of TiO_2 materials were also carried out in this study.

EXPERIMENTAL

Chemicals and Materials

Aniline (Sigma-Aldrich), Ammonium peroxydisulfate (APS) (Sigma-Aldrich, 98% reagent grade), Reactive Red 4 (RR4) dye (Sigma-Aldrich), Ethylene glycol (Sigma-Aldrich), Hydrochloric acid (HCl) (>37%, Merck), Titanium dioxide (TiO_2) (Degussa P-25, 80:20 of anatase: rutile), Commercially carbon doped TiO_2 (KRONOS, VLP-700), Cellulose filter paper (Whatman, Sigma-Aldrich), Acetone (Sigma-Aldrich), Methanol (Sigma-Aldrich) and distilled water.

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Methodology

The synthesis of emeraldine salt polyaniline (PANI) was according to the method by Rahmawati *et al.* [43]. In the first beaker, 9.8 g ammonium peroxydisulfate (APS) was dissolved in 100 mL distilled water. While in the second beaker, 3.72 g aniline was dissolved in 1 M of 100 mL hydrochloric acid. Both beakers were kept out in ethylene glycol bath at temperature 0℃ for 1h. APS solution was mixed with aniline solution and stored at temperature 0 ℃ for 24 h. The green precipitates formed were filtered by cellulose filter paper and further washed with acetone and 1 M hydrochloric acid solution.

For TiO_2 formulation preparation, 13 g of KRONOS TiO_2 is dissolved in 100 mL of distilled water and placed into an orbital shaker (PSU- 20i, Grant-bio, United Kingdom) for 30 min. The same procedure was repeated to study the effect other type of TiO_2 by using P25-TiO_2 and effect of dye sensitization by replacing 100 ml distilled water with 100 ml 30 mg L^{-1} reactive red 4 dye (RR4) for comparison study.

For PANI/TiO_2 solar cell preparation, two pieces of indium tin oxide (ITO) (2.5 x 2.0 cm) were cleaned using methanol and left to dry in the oven at 60 ^{o}C before use. PANI and TiO_2 were coated onto a different ITO glass plates by using a drop-casting method onto 4 cm^2 (2 x 2cm) and sandwich attached for the solar cell preparation. A paper clip is used to hold permanently the sandwiched solar cell.

The voltage was detected by using a compact digital multimeter (MT-1210, Pro'sKit, Taiwan) under 100 mW cm^{-1} of 250 W metal highlight (LIKO, Malaysia). Analysis of the functional group in the prepared PANI sample was done by FT-IR spectrometer (Nicolet380, Thermo Scientific, USA). X-ray diffraction (XRD) analysis (D5000, Bruker, Germany) was used to analyze the structure of the composite. Scanning electron microscope (JSM-6700F, JEOL, Japan) was used to study the morphology of the prepared sample. Electron lifetime was analyzed via electrochemical impedance spectroscopy (EIS) (10 mHz to 100 kHz) (ZIVE SP2, WonATech-ZIVE Lab, Korea).

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RESULTS AND DISCUSSION

Photovoltaic study

Table 1 shows the voltage production from PANI/TiO_2 sandwiched solar cell at different loading. The sandwiched solar cell that consists of 0.2: 0.2 g of PANI: TiO_2 ratio (PT4) is the optimum and the highest voltage recorded (0.66 V) among all prepared sandwiches solar cell. TiO_2 loading beyond 0.2 g has shown decreasing voltage production due to the less penetration of light and deficiency of free produced electron in TiO_2. In this case, the band gap energy of materials plays important role where the large band gap energy may reduce the photon energy absorption resulting the less transition of electron between valance to conduction band.

Meanwhile, the voltage values at different loading of PANI at 0.2g TiO_2 (PT7-PT11) showed a slightly comparable with optimum (PT4). This observation might be due to the maximum of free electron produces in TiO_2. As a result of high electron, the open circuit voltage and the photocurrent density are supposedly to be increased while the charge recombination is reduced [44-47]. PT7 shows no detection of voltage value, it indicates that without PANI, the system cannot be completely functioning. The free electron has only been captured by PANI at various ratio with the optimum of electron capture was 0.2:0.2 of PANI: TiO_2. The voltage values are sequentially increase for PT4>PT4'>PT25>PT25' as can be seen in Table 1. PT25 sample which containing P25-TiO_2 shows the lower voltage reading and this might be due to the large bandgap energy obtained from P25-TiO_2 make it less response on wide optical range [9,12].

Table 1: Voltage production from PANI/TiO_2 sandwiched solar cell at different loading under 100 Wm^{-2} of metal highlight

Sample	Ratio (g)			Voltage*
	PANI	Kronos TiO_2	Description	
PT1	0.20	0	-	0.00
PT2	0.20	0.05	-	0.39
PT3	0.20	0.10	-	0.55
PT4	0.20	0.20	-	0.66
PT5	0.20	0.30	-	0.58
PT7	0.00	0.20	-	0.00
PT8	0.05	0.20	-	0.59
PT9	0.10	0.20	-	0.59
PT4	0.20	0.20	-	0.66
PT11	0.30	0.20	-	0.60
PT4'	0.20	0.20	RR4 dye sensitized	0.64
PT25	0.20	0.20 (P25-TiO_2)	-	0.57
PT25'	0.20	0.20 (P25-TiO_2)	RR4 dye sensitized	0.55

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Characterization study

Figure 1 shows the FTIR spectrum of synthesized PANI powder in the 4000-500 cm^{-1} range. A major peak was observed at 1637.69 cm^{-1} presence the absorption and stretching of PVP [46]. The broad peak at 3333.40 cm^{-1} is correspond to O-H bond representing of moisture in the sample. It can be seen that the main band of pure PANI at 1475 cm^{-1} indicates the stretching of benzoid ring of C=N stretching mode. The band at 1211 cm^{-1} is assign to the stretching mode of C-N [47]. The band at 1157 cm^{-1} represent the N=Q=N, where Q is the quinonoid unit, while the band at 831 cm^{-1} indicate C-C and C-H of benzenoid unit.

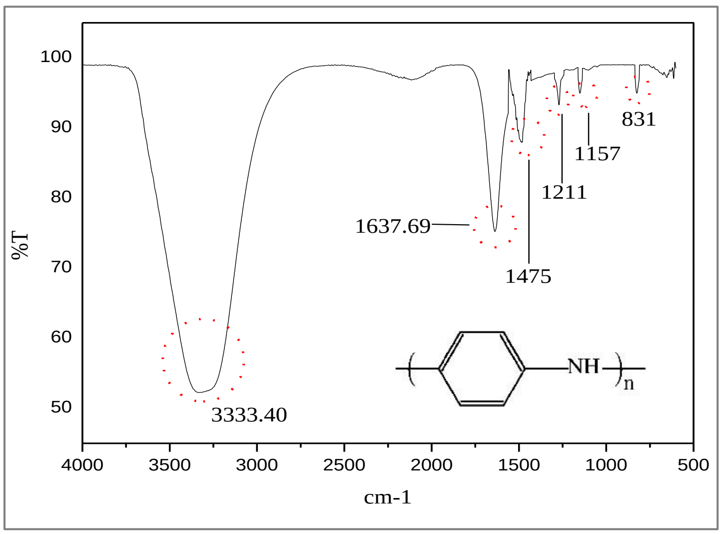

Figure 1: The FTIR spectrum for PANI powder and PANI molecular structure

X-ray diffraction (XRD) is used to study the crystallographic structures of the composite materials. The XRD pattern of PANI is shown in Figure 2. This spectrum was measured in a range of 2θ from 10° to 80°, showing strong and sharp peak at 25.99° correspond to (200) plane of polyaniline confirming the presence of emeraldine salt (ES) of PANI [48]. This also can be assigned to the scattering from PANI chains at interplanar spacing. Other two strong peaks also can be observed are at 30.03° and 24.21°, which show the characteristic of polyaniline [49-50]. The peak at 24.21° reveals that PANI has also some degree of crystallinity [51].

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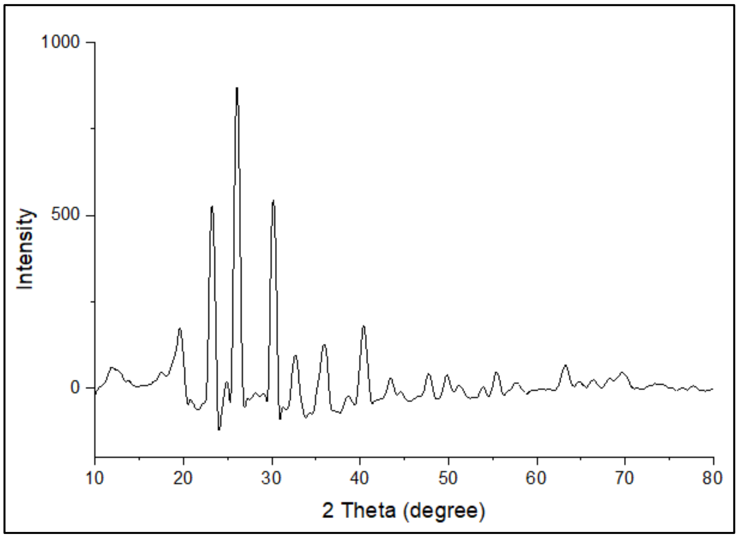

Figure 2: The XRD spectrum for PANI powder in a range of 2θ from 10 to 80° diffraction angle

The morphology of the surface of samples prepared is observed under 50K magnifications can be seen in the Figure 3 (a-c). The morphology of polyaniline can be seen in Figure 3 a) which PANI grow and agglomerate forming interconnected nanofiber network. Figure 3 b) shows the morphology of C doped TiO_2 with compact spherical particles agglomerates having porous irregular cauliflower-like features. The morphology of TiO_2 (P-25) can be seen in Figure 3 c) which shows formation of small clusters of spherical shape particles.

The typical Nyquist plot for electron lifetime (τ_e) of the different types of TiO_2 and RR4 sensitization (PT4, PT4’, PT25 and PT25’) are shown in Figure 4. From the Nyquist plot shown in Figure 2, the *fmax* for PT4, PT4’, PT25 and PT25’ are estimated to be 13.178, 10.3833, 6.6845 and 4.1508 Hz respectively. Hence, the electron lifetime (τ_e) of PT4, PT4’, PT25 and PT25’ are 20.76, 16.31, 10.56 and 6.52 s respectively. Therefore, based on the τ_e it can be conclude that TP4 have better photovoltaic performance. These results are in-line with the voltages reading in Table 1 which was employed for characterizing three prepared samples which are Polyaniline (PANI), TiO_2 and C doped TiO_2.

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a) b) c)

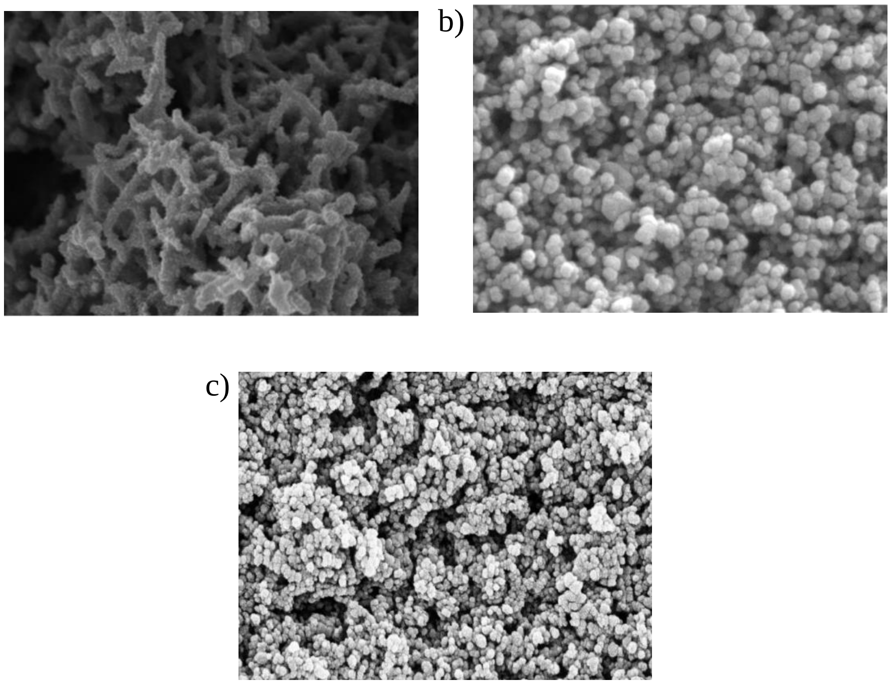

Figure 3: Surface morphology of (a) PANI (b) carbon-doped TiO_2, KRONOS and (c) TiO_2 (P-25) samples

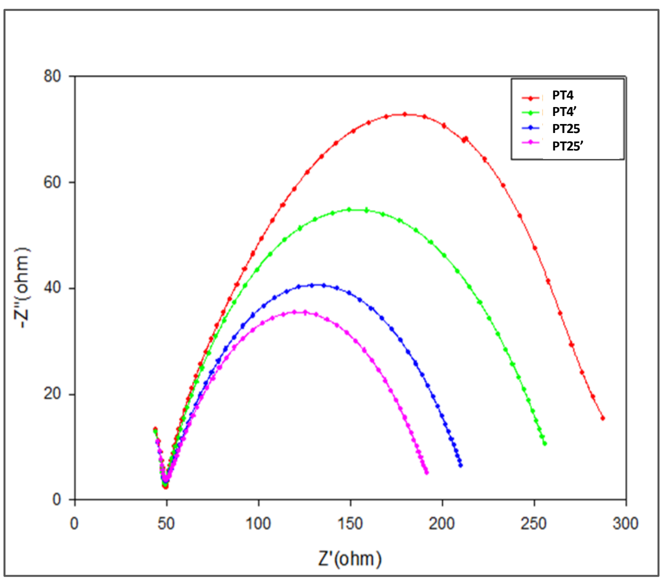

Figure 4: The Naquist plot of different type solar cells

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Stability study

The voltages reading produced upon days for PT4, PT4', PT25 and PT25' solar cells are shown in Figure 5 a). From the graph in Figure 5 a), it clearly seen that PT4 produces a higher voltage as compared to others solar cells. PT25 which is consist of different TiO_2 material (P25-TiO_2) shows a slightly lower voltage as compared with PT4 at first day and it's gradually decreased to become 0 voltage after 24 days. Despite of having higher voltage to others, PT4 has also shown a voltage stability upon the days was taken where the value is around 0.66 V ±0.03. This might be due to the complex structural of C incorporated in TiO_2 lattice structure in Kronos TiO_2 where it is chemically bonded forming Ti-C-O which give the significant effect on the sustainability for PT4 solar cell [52]. Due to C-doping in Kronos TiO_2, new energy states are produced deep in the TiO_2 band gap (substitution of oxygen by carbon atoms), which are in charge for the visible light absorption, [53].

However, the sustainability is becoming erupted where the voltage was drastically dropped to 0 volt in just only 24 days for PT4' due to the additional RR4 dye. Same goes to PT25' where the voltage become tremendously reduced. This might be due to the photo-oxidation of RR4 dye forming the intermediates which have eventually become poisoning to the photoexcitation of free electron [54]. Figure 5 b) shows the voltage reading for TP4 sample was recorded in every month and its look like the produced voltage is still the same at 0.65 V ± 0.03 for over 12 months. It shows the C in TiO_2 has play a significant role in sustaining the effectiveness in the system. Kronos TiO_2 used in this experiment consist substitutional C doped TiO_2 where it is chemically bonded with titania (Ti) forming a titania carbide (Ti-C). This makes Kronos TiO_2 (T4') has a synergistic effect of reducing the bandgap energy of the semiconductor and sustaining the photovoltaic at the same time.

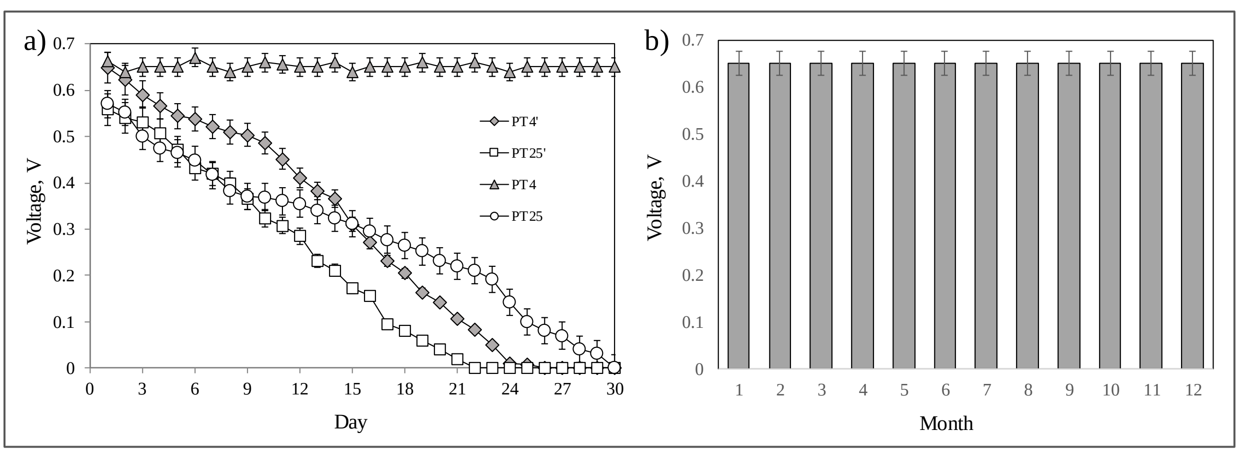

Figure 5: The voltage values for a) different solar cells approached up to 30 days and b) PT4 solar cell extended to 12 months

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CONCLUSION

PANI/TiO_2 solar cell has been successfully prepared by using sandwiched method. TP4 sample which containing 0.2, 0.2 g of PANI and TiO_2 loading coated on 2 cm^2 of ITO glass using drop casting method is the optimum PANI/TiO_2 solar cell by giving the highest voltage of 0.66 V. PANI with different TiO_2 has shown different behavior such as lifetime (τe), voltage and sustainability. No voltage reading was observed in PT7 sample, it shows the prepared solar cell system will not functioning without presence of PANI. Adding RR4 dye in solar cell sample will reduce the voltage by 13.7% to 0.57 V. It also drastically reduces the solar cell sustainability in which able to stand just for 30 days. As such, based on the sustainability of up to 12 months makes PANI/TiO_2 a bright potential for future solar cell.

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CONFLICT OF INTEREST STATEMENT

The authors hereby declare that there are no conflicts of interest whatsoever.

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