Effect of UV-Light Treatment on Efficiency of Perovskite Solar Cells (PSCs)

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Abstract: We employed ultra-violet (UV) light treatment on the TiO\textsubscript{2} layer prior to coating the perovskite layer to improve the solar conversion efficiency of perovskite solar cells (PSCs). A laboratory-made UV treatment system was equipped with various UV light sources (8 W power; maximum wavelengths of 254, 302, and 365 nm). The UV light treatment improved the power conversion efficiency (PCE) while coating the uniformity layer and removing impurities from the surface of cells. After the PSCs were exposed to UV light, their PCE developed approximately 10% efficiency; PBI\textsubscript{2} decreased without changing the structure.

Keywords: UV light; perovskite; solar cell; surface; treatment

1. Introduction

In recent years, renewable energy sources have attracted increasing attention due to oil depletion and other environmental problems. Several studies have been conducted to address the problems. Converting solar energy into heat, fuel, and electricity is one of the most effective ways of obtaining renewable energy [1]. Therefore, silicon-based solar cells now dominate the photovoltaic market due to the higher efficiencies of up to 25% compared to other solar cell devices. Owing to the high production cost of silicon-based solar cells, interest in organic material-based solar cell systems is increasing [2]. Compared to inorganic photovoltaic cells, organic material-based solar cells such as organic solar cells, dye-sensitized solar cells (DSSCs), and perovskite solar cells (PSCs) have low production costs, are lightweight, and have flexible device fabrication [3–6]. Despite their excellent advantages, organic material-based solar cells have low conversion efficiencies and unstable small-size devices (~1 cm\textsuperscript{2}) [7]. In the majority of works that have been reported, research groups have focused on higher efficiency in solar cell devices. The efficiency has been improved mainly by determining new perovskite material composition and modified film deposition processes [8].

Currently, PSCs have many layers, such as a transparent electrode (fluorine-doped thin oxide, FTO), a compact electron transport layer (ETL), a perovskite layer, a hole transport layer (HTL), and a metallic electrode (Au, Ag, and Cu). The active layers between two electrodes are coated by a two-step method. Despite performing the PSC fabrication process in a well-controlled glove box under low-level humidity, moisture, and oxygen, defects and poor adhesion between layers appeared [9,10]. Moreover, after a compact TiO\textsubscript{2} layer and a mesoporous TiO\textsubscript{2} layer were deposited on the FTO electrode, the temperature increased to over 450 °C. Therefore, to achieve a high energy conversion performance of PSCs, the layers should be smooth and uniform [11–13].

Ultraviolet (UV) irradiation has been mainly used for photocatalytic applications on the surface or for improving the surface coating energy. UV light is classified as A (400–315 nm), B (315–280 nm), and C (280–200 nm). UV radiation has shorter wavelengths than visible light. Unfortunately, all UV light is not helpful to humans, and UV-A and UV-B are related to skin cancer [14]. Among them, UV-C
is used to remove impurities on the surface by induced ozone (O₃) in the air. Some research groups have tried to enhance the power conversion efficiency (PCE) of solar cells [15,16]. Until now, other surface science fields have reported on whether oxide materials, such as TiO₂, SnO₂, and ZnO, have photocatalysis effects in a range of light with various wavelengths [17,18]. Work comparing various ultraviolet light has not yet been conducted in the field of PSCs. Moreover, based on previous studies, we confirmed that UV light irradiation (UV-C) treatment improved the surface of a dye-sensitized solar cell (DSSC) [19]. However, there have been very few studies on other UV light sources (A and B).

In this study, we investigated the effect of the UV light source and the light irradiation time of the PSC fabrication process to enhance its PCE.

2. Experimental Details and Measurements

2.1. Materials and Preparation of Solar Cells

Chemical reagents were purchased from Sigma-Aldrich chemical company and were used without further purification. The two-step spin-coating method was performed for the PSC layer and thermal evaporation for the Ag and Au electrode (Kojundo Co. Ltd, 99.95%). The FTO substrate (thickness < 2.2 mm, sheet resistance < 8 ohm/sq) was purchased from Wooyang GMS Company. Prior to coating the chemical solutions, we cleaned the FTO substrate with acetone, ethanol, and deionized water using ultrasonication. The substrates were then blown by nitrogen gas to remove any remaining impurities or contamination. Three kinds of solutions were required to form the PSC layer—TiO₂ paste, CH₃NH₃PbI₃, and spirobifluorene (Spiro-MeOTAD). First, laboratory-made TiO₂ paste was used to form a compact layer (C-L) and a mesoporous layer (M-L). The TiO₂ paste was prepared with TiO₂ powder (anatase < 25 nm, 99.7%, Sigma-Aldrich). Second, CH₃NH₃PbI₃ was formed during the spin-coating process with CH₃NH₃I and PbI₂ solutions, according to a previous study [20]. Finally, a spiro-MeOTAD solution was dissolved in a mixture of chlorobenzene, 4-tert-butyl pyridine and lithium bis(trifluoromethanesulfonyl)imide (Li-TFSI) (520 mg Li-TSFI in 1 ml acetonitrile, Sigma-Aldrich) and used without any additional processing.

The schematics of the PCS fabrication procedure are shown in Figure 1. Prior to the fabrication of the solar cells, fluorine-doped tin oxide (FTO)-coated glass substrates (sheet resistance: 8 ohm sq) were purchased from Wooyang GMS Company. Prior to coating the chemical solutions, we cleaned the FTO substrate with acetone, ethanol, and de-ionized water in a bath-type sonicator for 20 min each. Any impurities on the cleaned substrates were removed by nitrogen gas before drying in an oven at 100 °C for 30 min.

![Figure 1. Schematics of the perovskite solar cell (PSC) fabrication procedure with ultraviolet (UV) light treatment (DL-S = distance from light to sample).](image)

The PSC fabrication procedures were carried out in a nitrogen gas filled groove box. The compact TiO₂ (c-TiO₂) layer was coated at 3000 rpm for 30 s at 120 °C for 10 min on a hot-plate, separately. Then, the mesoporous TiO₂ (m-TiO₂) layer was deposited on the c-TiO₂-coated FTO substrate at 3000 rpm for 30 s at 450 °C for 10 min in a furnace. The CH₃NH₃PbI₃ layer was formed by combining CH₃NH₃I and PbI₂ solutions on the m-TiO₂/c-TiO₂ film by the spin-coating method [21–23]. After the spiro-MeOTAD layer was coated at 4000 rpm for 30 s, the samples were loaded on a thermal evaporator.
machine. Approximately 100 nm of Ag was thermally evaporated on the spiro-MeOTAD-coated film in a vacuum (under $10^{-6}$ torr).

2.2. UV Surface Treatment of Samples

Laboratory-made UV irradiation equipment consisted of an aluminum reflector and 8 W UV light lamps with maximum wavelengths of 254, 302, and 365 nm. More detailed equipment and setup conditions are shown in Figure 1. Prior to depositing the CH$_3$NH$_3$PbI$_3$ layer on the m-TiO$_2$/c-TiO$_2$-coated FTO substrate (sample size: 20 mm × 20 mm), the UV light treatment was carried out on the surface at room temperature in a sealed box.

2.3. Measurements

The structure and morphology of the prepared samples were observed using field-emission scanning electron microscopy (FE-SEM, Hitachi S-4500) and X-ray diffraction (XRD, Rigaku corporation SmartLab) using Cu-Kα radiation ($\lambda = 1.54056$ Å, 9 kW) in the $2\theta$ range of 20–80°. The contact angles of the water droplets on the solid surface were measured using a contact angle meter. The conversion efficiencies of the prepared solar cell devices were measured using a current–voltage (J-V) solar simulator (K3400, McScience Company) adjusted to AM 1.5G (100 mW/cm$^2$).

3. Results and Discussion

Firstly, to investigate whether UV treatment on the film surface influenced the structural properties and surface morphology, we measured the water contact angle (WCA) of an m-TiO$_2$/c-TiO$_2$/FTO substrate and images of samples coated with the CH$_3$NH$_3$PbI$_3$ layer, as shown in Figure 2. The samples were treated for 1 h by UV light A, B, and C, separately. As shown in Figure 2, compared to the reference sample without surface treatment (approximately 20°), the WCA of all samples with UV light surface treatment decreased due to the hydrophilic surface of the substrate. Among them, it was thought that UV-C light was the most effective to obtain a high-energy, for example, hydrophilic surface [24]. As light has a longer wavelength, transmittance increased and energy was low. It is assumed that developing surfaces have more effect.

![Figure 2](https://example.com/figure2.png)

**Figure 2.** Water contact angle (WCA) as a function of the UV light wavelength (A, B, or C); (a) without UV light treatment, (b) 254 nm, (c) 302 nm, and (d) 365 nm.
Therefore, we selected UV-C light with the shortest wavelength and investigated the properties of samples treated under other conditions. Subsequently, we reduced 60 min to 30 min, and the change in the surface wettability of the film surface was confirmed by the WCA, as shown in Figure 3a,b. Despite the reduction of the surface treatment time, we were able to obtain a hydrophilic surface. The median of three measurements of the WCA values was calculated as the median of three determinations on recorded images of the samples. Following UV-C treatment of the m-TiO2/c-TiO2/FTO substrate, the WCA of the surface decreased from 17° to approximately 0°. The UV-C light from specific wavelength ranges was introduced to enhance the surface energy, and its surface temporarily became hydrophilic. Moreover, owing to the enhanced hydrophilicity of the sample surface and the strong hydrogen bonds to water molecules, the dye–liquid solution could uniformly cover the m-TiO2/c-TiO2/FTO substrate in the spin coating process after the liquid solutions were dropped on the sample [25–27].

![Figure 3a](image1.png) ![Figure 3b](image2.png)

**Figure 3.** (a,b) Water contact angle (WCA) measurements of m-TiO2/c-TiO2/FTO substrate; (c,d) photo images of samples coated with the CH₃NH₃PbI₃ layer; (a,c) before UV-C light treatment; and (b,d) after UV-C light surface treatment.

We observed the sample surface after dropping the CH₃NH₃PbI₃ solution onto it and the photo images of the samples are shown in Figure 3c,d. After the m-TiO2/c-TiO2/FTO substrate was exposed to UV-C light, we obtained samples with better film-coating uniformity. Further, the sample had no defects unlike samples without UV-C light surface treatment (film defects: dotted lines in samples, as shown in Figure 3d).

We investigated the effect of the crystal structure on the UV light surface treatment. Figure 4a, which displays the XRD patterns of the m-TiO2/c-TiO2/FTO substrate, shows the variation in crystallinity before and after the UV light surface treatment. The XRD peaks of all samples were detected in the range of 10–80°. All strong diffraction peaks of the samples matched with the standard TiO2 peaks (ICDD card no. 03-065-5714). No structural shift was observed following the UV treatment. The XRD peaks of 24.4°, 48.2°, 62.8°, and 75.2° represented the reflections from the (101), (004), (200), and (211) crystal plane of an anatase structure, respectively.

![Figure 4a](image3.png)

Figure 4b presents a cross-sectional SEM image of fabricated PSCs after UV treatment. The m-TiO2/c-TiO2/FTO (500 nm), CH₃NH₃PbI₃ (200 nm), Spiro-MeOTAD (100 nm), and Ag electrode
(100 nm) were sequentially stacked on the substrate as shown in Figure 4b. The thickness of the layers was consistent with the configuration previously reported in a similar fabrication procedure [20].

Figure 4. (a) X-ray diffraction (XRD) patterns of m-TiO$_2$/c-TiO$_2$/FTO substrate and (b) cross-sectional field-emission scanning electron microscopy (FE-SEM) image of fabricated perovskite solar cells (PCSs) after UV-C light surface treatment.

The optical absorbance spectra of coated PbI$_2$ are shown in Figure 5. After the UV-C treatment of the sample, the height of the absorbance peak of PbI$_2$ detected at around 481 nm decreased. The UV-C treatment was conducted by a combination of two wavelengths, i.e., 84 and 254 nm. 1) Ozone (O$_3$) gas was generated from oxygen in the air by light irradiation at 184 nm. 2) Impurity particles such as organic contaminants attached to the surface were removed by oxidation with O$_3$ after irradiation by light at 254 nm. Furthermore, Saekow et al. [28] reported that after UV-C treatment, the light could generate O atoms as well as OH$^-$ groups from the H$_2$O molecules in the air. In addition, the O atoms on the surface of the TiO$_2$ layer were replaced by OH$^-$ groups. Owing to an increase in the number of O$^-$ and O$^{2-}$ species, more Ti$^{3+}$ ions were produced compared with Ti$^{4+}$ ions. The increase in the
number of Ti$^{3+}$ ions on the surface could attract and combine the carboxylic groups with the PbI$_2$, and the PbI$_2$ concentration decreased as the absorbance peak of PbI$_2$ was reduced, as shown in Figure 5.

Figure 5. Absorbance of PbI$_2$-coated m-TiO$_2$/c-TiO$_2$/FTO substrate with and without UV-C light surface treatment.

We prepared 4 reference samples (without UV light surface treatment) and 18 UV-light treated samples (UV-A, UV-B, and UV-C). We used Ag as the metal electrode. When the wavelength was shorter, the efficiency of PSCs improved, as shown in Figure 6a.

Figure 6. (a) Efficiency and (b) External quantum efficiency (EQE) of perovskite solar cells (PSCs) treated by UV light sources (A, B, and C).

As can be seen from the EQE data of Figure 6b, the longer wavelength of the UV light source resulted in smaller quantum efficiency, which is expected to be the result of the recombination at the surface. A short-wavelength indicates that the grain size does not increase significantly but that the roughness increases resulting in a lowered fill factor. Longer the wavelengths indicated lower efficiency due to the decrease in the current density. Among the samples with UV surface treatment, the efficiency of that with UV-C treatment was the highest. It was confirmed that a UV-C light sources have the greatest effect on improving the performance of PSCs. Therefore, we investigated the effect of light exposure time on PSCs.

UV light at 245 nm was irradiated onto the TiO$_2$ layer on the FTO substrate for 10, 20, and 30 min. An absorbance graph, Figure 7 was obtained, and the absorbance of the whole region, except the PbI$_2$ peak, was further increased compared with the conventional PbI$_2$ peak. The TiO$_2$ layer is
cleaned by UV-C light irradiation and it has an appropriate structure, which could be allowed to be better perovskite formation on the surface. Moreover, the formation of perovskite, which enables well bonded at the perovskite/TiO₂ interface, normally transfers electrons to the TiO₂ layer, disturbing the open-circuit voltage (VOC). The EQE measurements were employed to provide independent corroboration of the short-circuit current-density (JSC) values collected with the solar simulator. The most significant influence on PCE was given by improvement in the JSC values, as a consequence of overall improved EQE behavior for all samples.

**Figure 7.** Photovoltaic performances of perovskite solar cells (PSCs) as a function of the UV-C light irradiation time; (a) open-circuit voltage, (b) short-circuit current, (c) short-circuit current-density, (d) fill factor, (e) power conversion efficiency (PCE) of prepared PSC device, and (f) External quantum efficiency (EQE). The box plots of (a)–(e) were drawn vertically indicating data distribution of photovoltaic parameters recorded from a solar simulator (AM 1.5 AM 1.5G illumination). The minimum, maximum, and average values are shown by the middle line of each box.
The UV-C light exposure time increased the efficiency of the manufactured solar cell when the perovskite was applied with the $V_{OC}$ reduction, as confirmed in Table 1.

**Table 1.** Average and standard deviation photovoltaic performance of solar cells with various UV-C treatment times.

| Parameters                        | $^a V_{OC}$ (V) | $^b I_{SC}$ (mA) | $^c J_{SC}$ (mA/cm$^2$) | $^d F.F.$ (%) | $^e PCE$ (%) |
|-----------------------------------|----------------|-----------------|------------------------|--------------|-------------|
| Without UV-C treatment            | 0.953          | 1.284           | 24.239                 | 61.37        | 14.142      |
|                                   | (0.0436)       | (0.0789)        | (1.489)                | (2.080)      | (0.232)     |
| With UV-C treatment               |                |                 |                        |              |             |
| 10 min                            | 1.008          | 1.229           | 23.198                 | 64.602       | 15.076      |
|                                   | (0.045)        | (0.077)         | (1.457)                | (2.165)      | (0.288)     |
| 20 min                            | 0.942          | 1.260           | 23.774                 | 63.312       | 14.794      |
|                                   | (0.030)        | (0.048)         | (0.921)                | (2.325)      | (0.223)     |
| 30 min                            | 0.961          | 1.302           | 24.573                 | 61.537       | 14.493      |
|                                   | (0.044)        | (0.058)         | (1.110)                | (3.169)      | (10.745)    |

$^a$ Open-circuit voltage, $^b$ Short-circuit current, $^c$ Short-circuit current-density, $^d$ Fill Factor, and $^e$ Power conversion efficiency.

The $V_{OC}$ of the UV-C treated perovskite solar cell increased from 0.953 to 1.008 and the mean efficiency increased from 14.142% to 15.076%.

Figure 8a shows the J-V characteristics of the fabricated cells after UV treatment under AM 1.5G illumination (100 mW/cm$^2$). Details of the device parameter values measured from the solar stimulator are listed in Table 1. The PSCs without UV radiation had a $J_{SC}$ of 24.239 mA/cm$^2$, an F.F. of 61.370%, a $V_{OC}$ of 1.284 V, and a PCE of 14.142%. The PSC samples exposed to UV light had a $J_{SC}$ of 24.239 mA/cm$^2$, an F.F. of 64.602%, a $V_{OC}$ of 1.008 V, and a PCE of 15.076%. The improvement in the values was caused by the variation of $J_{SC}$.

The parameter $J_{SC}$ was attributed to the number of photo-generated electrons transferred to the perovskite layer through the TiO$_2$ layer. Therefore, it is important to improve the photovoltaic conversion efficiency of PSCs to improve the interfacial properties between TiO$_2$ and perovskite layers. The TiO$_2$ film surface irradiated by the internal resistance and charge transfer kinetics of prepared PSCs was measured using an electrochemical impedance spectroscopy (EIS) analyzer in light–dark conditions [29,30].

Figure 8b shows the EIS results of the samples prepared under the condition of UV-light irradiation. The diameter of the second semicircle is a measure of the recombination resistance ($R_{rec}$) in the PSC samples, which systematically increased after UV-light exposure, thereby indicating that the UV treatment was characterized by an increased recombination of UV light irradiation compared to no light-exposure. Higher RCT was induced for lower current loss and an increased F.F.

To confirm the effect of UV-C treatment on the PCSs, seven samples were selected, and their average PCE was calculated. After UV treatment, their maximum efficiency increased by 14.026% to 15.583%, as shown in Figure 8a.
prepared PSCs was measured using an electrochemical impedance spectroscopy (EIS) analyzer in light–dark conditions [29,30].

Figure 8. Photovoltaic properties of perovskite solar cells (PSCs) before and after UV treatment; (a) current–density and (b) Nyquist plots of PSC device. The size of the box plot interquartile is the third quartile (Q3) to the first quartile (Q1).

4. Conclusions

In this study, we investigated the effect of UV-light source and irradiation time on the fabrication of PCSs. Their power energy conversion efficiency was investigated for a simulated solar irradiation power density of 100 mW/cm² (AM 1.5G). After UV treatment on the c-TiO/m-TiO₂/FTO substrate, the film surface became hydrophilic; the perovskite layer was easily and uniformly deposited on the film. The PCE of PSCs treated by UV-C light increased more than those treated with UV-A or UV-B. Samples irradiated with UV light had a low series resistance, high recombination resistance inside the device, and a high PCE of approximately 10%, compared with PSCs without UV-light irradiation.

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