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Improving the Stability of the Carbon-Based Perovskite Solar Cells by a TiO_2 Templated Film with Self-Cleaning Property

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ABSTRACT

Perovskite solar cells (PSCs) have garnered significant attention due to their high efficiency and low-cost fabrication. However, their stability under environmental conditions, particularly UV exposure, remains a critical challenge. In this study, we address this issue by introducing a mesoporous TiO_2 templated film coated on the glass side of the FTO substrate, serving as both a UV absorber and a self-cleaning layer. The TiO_2 film was synthesized via a dip-coating method using a p123 copolymer as a template agent, resulting in a highly transparent and mesoporous structure. The self-cleaning properties of the film were evaluated through the degradation of Congo red dye under optimized conditions, including pH, dye concentration, and film thickness. The TiO_2 film demonstrated excellent photocatalytic activity and effectively degraded organic pollutants under UV irradiation, which highlights its potential for real-world applications. Furthermore, incorporating this film on the glass side of the carbon-based perovskite solar cells significantly improved their stability. This work presents a promising approach to enhancing the durability and performance of PSCs by integrating multifunctional TiO_2 films, paving the way for more robust and sustainable solar energy technologies.

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1. Introduction

In recent years, environmental pollution from fossil fuels has become a problem. Therefore, humans must change their approach to utilizing energy from fossil fuels to clean energy. Therefore, the use of renewable and clean energy sources such as solar energy has increased.

The sun, as a unique source, can provide the annual energy needs of the entire world in one hour. Solar energy is a large source of energy, having endless availability and cleanliness. In 1946, Russell Ohl made the first photovoltaic junction solar cell by using silicon to convert solar energy. [1] To date, numerous kinds of solar cells, such as multi-junctions, organics, perovskites, and many more, have been produced. Among these, in recent years, perovskite solar cells have attracted a lot of attention due to their low cost and the rapid growth of their efficiency [2]. A conventional perovskite solar cell consists of Glass/FTO/compact-TiO_2/mesoporous-TiO2/Perovskite/Spiro ometad/Au. The principal challenge for perovskite cells is their instability under environmental conditions like humidity and UV light. To increase the stability of the perovskite solar cells against humidity, carbon-based perovskite solar cells were introduced with 6.6% efficiency, [3]and today it has reached 20%. [4]

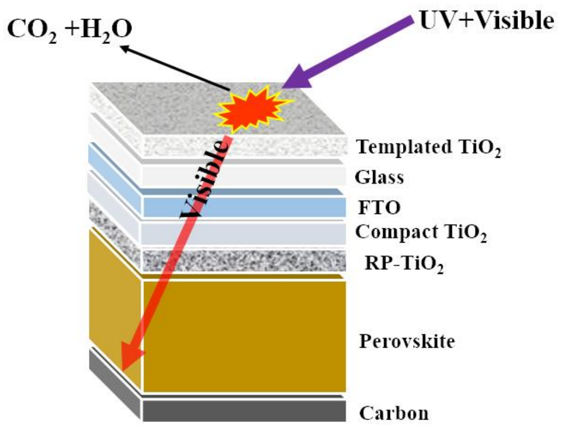
The TiO_2 electron transport layer used in the conventional PSC configuration contributes to the photocatalytic degradation of perovskite in the presence of ultraviolet radiation and therefore causes instability in the presence of ultraviolet light[5]. The photocatalytic degradation of perovskite in the presence of ultraviolet radiation occurs in the TiO_2 electron transport layer used in the conventional PSC configuration and therefore causes instability of these cells in the presence of ultraviolet.[6, 7] A common solution to protect perovskites from degradation in the presence of ultraviolet light entering the solar cell structure is to coat the cell's glass substrate on the outside surface with ultraviolet-absorbing layers. In addition to ultraviolet absorption, this structure coating must also have an anti-reflective feature. Most UV absorbers have a higher refractive index than glass, which increases the reflection of light and decreases the light entering the perovskite active layer, and therefore reduces efficiency. Porosity making is one of the simple ways to decrease the refractive index of the coated UV absorber materials and increase the input light transmittance through the glass substrate. [8] Mesoporous templated titania increases the porosity and input light transmittance, and could also be used as a transparent UV absorber material and destroy contaminants as a self-cleaning coating. [8] [9] However, if mesoporous titanium dioxide is placed outside of the cell (on the glass part of the substrate) as well, it captures UV light and improves the stability of the PSC. On the other hand, the external titanium dioxide destroys the contaminants placed on the glass surface of the perovskite solar cells by the photocatalytic effect and plays a self-cleaning role. This self-cleaning effect helps maintain cell efficiency over time.

One of the most important applications of mesoporous titania is the photodegradation of various environmental organic pollutants such as azo-dyes[10, 11] [12].

TiO_2 particles fixed on support (TiO_2 films) have many features, including the lack of requirements for separation of photocatalysts by some techniques, such as centrifuging. Also, the porous films of TiO_2 may be more effective than the compact films when the reactants are adsorbed in the photodegradation reaction on the surface of the catalyst. [13-15].

A variety of polymer templating techniques have successfully been used in the sol-gel preparation of mesoporous TiO_2 thin films using commercially available Pluronic F127, Pluronic P123, and Brij-56[16]. In this regard, pluronic copolymers are used as a template and, after the growth of the material, are removed from the inorganic hybrid film for creating pores and mesoporous structures. The use of copolymers in synthesizing TiO_2 film helps in controlling the mesoporosity, transparency, surface roughness, and hydrophilicity of the film [17, 18]. The mesoporous templated films, due to the higher surface area, adsorb higher amounts of organic pollutants and increase photocatalytic activity. Furthermore, the thicker films present a higher percentage of porosity and higher roughness. So the thickness of the films seems to influence the photocatalytic activity.

In this work, we used a mesoporous templated titanium dioxide transparent film coated on the glass side of the FTO substrate (outside of the cell) as a UV absorber and photocatalytic self-cleaning material. This film was prepared by a dip coating method from a solution of TiO_2 containing p123 copolymer as a templating agent. Then, the degradation of Congo red dye as an organic pollutant in samples was investigated to evaluate the self-cleaning effect of the film. Finally, the efficiency and stability of carbon-based perovskite solar cells containing this TiO_2-templated film were evaluated (Scheme 1).



Scheme 1. Schematic illustration of the photocatalytic self-cleaning process of mesoporous templated TiO_2 coated on the glass side of the PSC.

Experimental Section

Materials

Titanium (IV) isopropoxide (Merck,7.91g), ethanol (Aldrich), butanol (Aldrich,18.85), pluronic p123(2g), and Hcl (Merck, 36% wt, 4.85g) were used with no further purification to prepare titania templated solution.

Coating the mesoporous templated TiO_2 film on the glass side of the FTO

Accordingly, this section will explain the synthesis method of titanium dioxide sol first. Then, the glass side of FTO is washed, and the layer is deposited. Then, the mesoporous titanium is deposited using the dip-coating method.

Preparation of TiO_2 sol

First, 18.85 g of 1-butanol solvent was added to 2 g of p123 copolymer and then stirred for 1.5 hours. In another beaker, 4.85 g of hydrochloric acid was slowly added to the titanium isopropoxide. Finally, the HCl/$Ti(ipp)_4$ solution was added to the p123/butanol solution and aged by stirring without temperature for at least 3 h. The prepared TiO_2 sol was ready for deposition.[19].

Cleaning the glass

Before preparing thin films, the FTO glass must be free of contamination and dust. For this reason, the FTO glasses were washed in several steps. First, the FTO glasses were washed with soap and water in the ultrasonic bath. Then, they were washed with a distilled water solution, ethanol, and isopropanol for 15 minutes, respectively. After each washing step, the samples were dried with gentle air from a hair dryer.

Deposition of Mesoporous templated TiO_2 film

After the synthesis of titanium dioxide sol, the sol was used to synthesize the mesoporous TiO_2 layer using the dip-coating technique with a 30 cm/min−1 speed. After each dip-coating step, the FTO side was cleaned with ethanol so that only the glass side of the FTO was covered with TiO_2 film and then calcinated at 300 ºC for 15 minutes on a hot plate. After coating three layers, the layers were calcined at 450 ºC for 90min (heating ramp rate: 1 ºC/min) to increase the crystallinity and porosity of the mesoporous TiO_2 films. This method was repeated several times to obtain thicker layers (1 to 9 layers).

Coating on the FTO (Fabrication of carbon-based perovskite solar cells)

In this section, we will discuss the fabrication of a carbon-based perovskite solar cell. First, dense titanium dioxide is deposited on the glass side of FTO, which is coated with mesoporous titanium dioxide. Then, the same mesoporous titanium sol that was prepared in the previous step is used to deposit mesoporous titanium on dense titanium. After that, perovskite is deposited on the mesoporous titanium using the spin deposition method. Finally, carbon was fixed on the perovskite by the Dr. Blade method.

Preparation of TiO_2-dense layer

To synthesize dense TiO_2, 400 microliters of acetyl acetone were added to 9 ml of pure ethanol solvent and mixed well. Next, 600 microliters of titanium (IV) isopropoxide were slowly added to the solution. Simultaneously with the preparation of the TiO_2-dense solution, the covered glasses (3 mm with aluminum foil) were placed in the spray coating system. The temperature was set to 450 ºC, and the materials were sprayed onto the FTO from the nozzle. Finally, the temperature of the spray coating hot plate was set to 510 ºC, and the glasses were kept at this temperature for 30 min.

Preparation of mesoporous TiO_2-templated layer

The mesoporous TiO_2 solution was synthesized by dissolving block copolymer Pluronic P123 (Sigma-Aldrich, 1.0 g) in 1-butanol (9.07, Aldrich). To the tetraethyl orthotitanate (3.17 g, Sigma-Aldrich) on another baker, 2.42 g of HCl (Merck, 36 wt%) was added during vigorous stirring. A copolymer solution was added to the $HCl/Ti(OEt)_4$ solution after thirty minutes of stirring, and the solution was stirred continuously for three hours to create a clear TiO_2 solution.

$CH_3NH_3PbI_3$ perovskite preparation

First, 0.63 g of lead iodide and 0.21 g of methyl ammonium iodide were placed in an oven at 100 ºC for 2 hours to remove their water. To prepare perovskite, while the beaker was placed on a heater at a temperature of 75 to 80 ºC (without a stirrer), 1000 µl of DMF and 95.72 µl of DMSO were added to the lead iodide to dissolve at the temperature. Then, methyl ammonium iodide was added to it. The perovskite layer was deposited on the glass using the spin deposition method, in which the glass was placed in the center of the device from the FTO side, and about 50 µL of the prepared solution was poured onto it. The device was set at a speed of 4000 rpm. After 10 seconds, 200 µL of chlorobenzene was carefully added to the rotating glass. Then, the glass was placed on a heater at 100 degrees for 3 minutes[20, 21]

Conductive carbon paste deposition

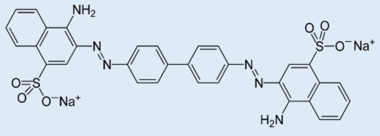
Conductive carbon paste) Dycotec, DM-CAP-4703S) was deposited onto the perovskite surface with the Dr. Blade method.

Preparation of Congo red solution

Finally, a fabricated carbon-based perovskite solar cell with a photoactive backside (titanium in mesoporous oxide) is used for dye degradation. First, a solution of a certain concentration of Congo red is prepared, and then the deposited FTO layer is placed in the solution.

To investigate the photocatalytic activity of mesoporous TiO_2 films, a 5 mg/l aquous dye solution of congo red (4-amino-3-[4-[4-(1-amino-4-sulfonato-naphthalen-2yl) diazenylphenyl] phenyl] diazenyl-naphthalene-1-sulfonate) was used. The additional information is shown in a table 1.

Table 1. Chemical properties of congo red organic compounds (AZO dye) used in dye degradation

λmax	Molecular weight (gr)	Chemical formula
490	696/665	

Characterization

A JASCO V-670 spectrophotometer was used to measure absorption spectra on solid samples. X-ray diffraction (XRD) measurements were carried out using a Bruker D8 Advanced instrument. The TiO_2 texture were studied by field emission scanning electron microscopy (FE-SEM, S4160 Hitachi Japan).

Results and Discussion

The X-ray diffraction pattern (XRD) of TiO_2 film (9-layer) coated on the glass side of the FTO substrate is shown in Figure 1. The main diffraction peak of anatase TiO_2 was observed at $2\theta = 25.24^\circ$, corresponding to the (101) crystal plane. The other less intense diffraction peaks of (200), (105), (211), and (204) corresponded to crystal planes of the anatase phase, and were marked on the XRD pattern [22].

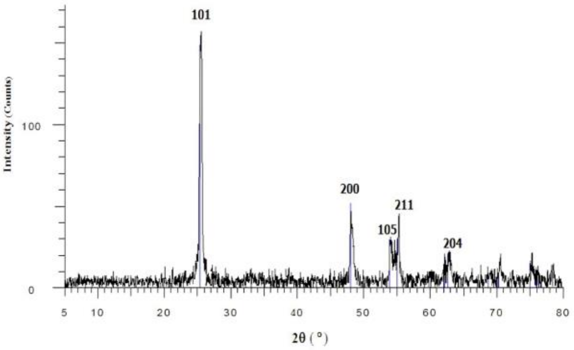


Figure 1. X-ray diffraction pattern (XRD) of mesoporous TiO_2 templated film (9-layer) coated on the glass substrate.

Figure 2 shows an FE-SEM image of mesoporous templated TiO_2 film with a worm-like morphology with open pores structures and without any crack on the surface.

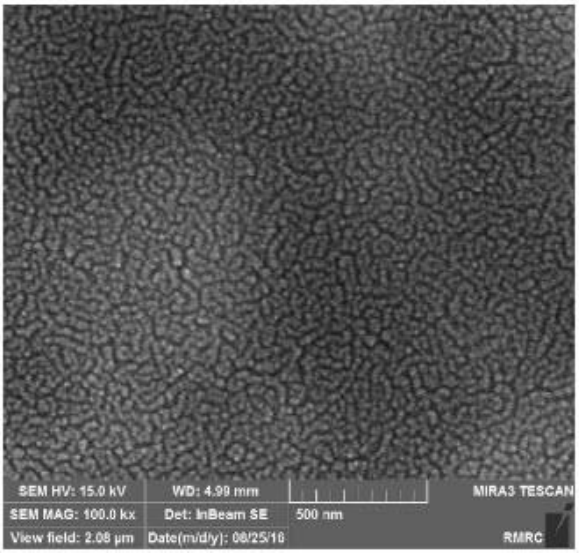


Figure 2. FE-SEM image of mesoporous TiO_2 films on the glass side of the FTO.

The purpose of using a titanium dioxide layer on the outer part of a carbon-based perovskite solar cell, or in other words, the glass part of the FTO substrate of the cell, is that it can both absorb ultraviolet radiation and have a photocatalytic self-cleaning property and also helps PSC stability.

The photocatalytic activity of films is influenced by various factors such as the nature and structural properties of the film, the solution acidity, the nature of the degraded organic compounds, and their initial concentration in the solution.

To investigate the effect of thin film thickness on the amount of azo dye adsorption, the film samples were placed in the dark for two hours in the presence of the photocatalyst. As shown in Figure 3, as the number of layers increases to 9, the titanium dioxide thin film adsorbs more dye. This can be due to increased porosity, specific surface area, and active layer thickness. It is worth noting that, the coating number of layers raises to 12 layers but due to the cracking of the layers and the decrease in transparency, the coating was not increased more than 9 layers.

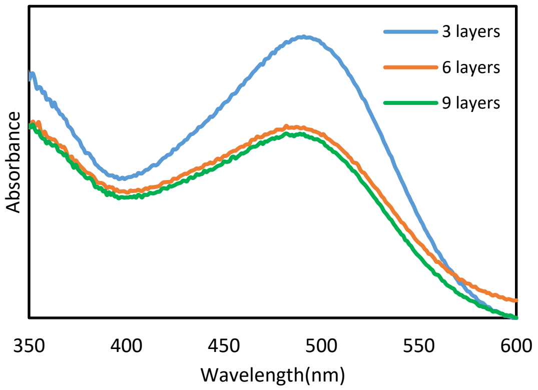


Figure 3. Absorption spectrum of Congo Red (concentration of 5 ppm) solution after adsorption on the mesoporous TiO_2 templated films with thicknesses of 3, 6, and 9 layers annealed at 450 °C for 5 h.

To examine the effect of ultraviolet light and mesoporous TiO_2 photocatalysts on the decomposition of the dye, first, the dye was exposed to ultraviolet light for two hours without the presence of photocatalysts. The results showed that the ultraviolet light source could not effectively degrade dye in the absence of a photocatalyst. Then, the mesoporous TiO_2 films were exposed to the dye solution for two hours without ultraviolet light in the dark. In this case, no significant removal of Congo red was observed. In these conditions, the rapid achievement of an equilibrium between dyes and the film surface can be the cause of slight changes in the dye concentration. When the dye degradation is performed under ultraviolet light and TiO_2-templated film, the total dye removal is significantly increased so that the surface of the films is also transparent and not red. All dye degradation experiments were started after two hours of dye contact with the films in the dark to ensure a proper balance between the dye and the film surface. The UV-Vis spectrum of the dye with a concentration of 15 ppm during the degradation reaction using a mesoporous TiO_2 film under ultraviolet light is shown in Figure 4.

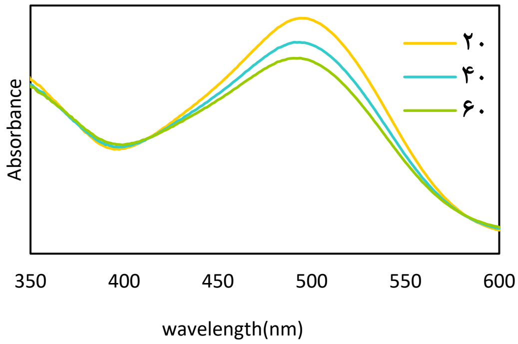


Figure 4. UV-vis absorbance spectrum of Congo red dye at a concentration of 15 ppm during the degradation reaction at different times (20, 40, and 60 min) using a TiO_2-templated photocatalyst under ultraviolet light.

Moreover, in the presence of 9-layer mesoporous titanium dioxide, Congo red degradation with different concentrations (5 ppm, 10 ppm, and 15 ppm) over time was investigated (Figure 5, Figure 6, and Table 2). The lower the dye concentration, the higher the adsorption rate, and the more degradation operations will be performed over time. It should be noted that the photocatalytic activity of dye degradation for a concentration of 5 ppm continued for 100 minutes for a complete reaction.

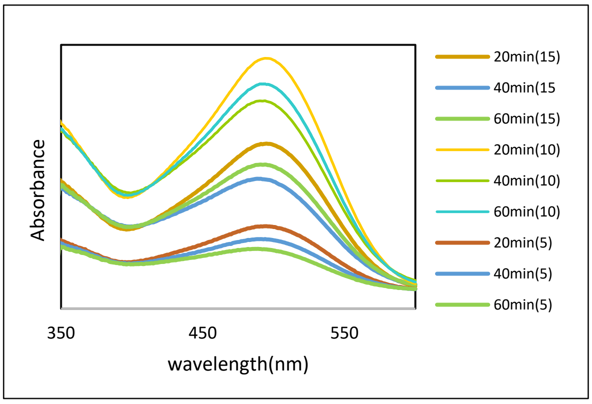


Figure 5. UV-vis absorbance spectra showing the Congo red degradation with different concentrations (5 ppm, 10 ppm, and 15 ppm) over time in the presence of 9-layer mesoporous templated titania film

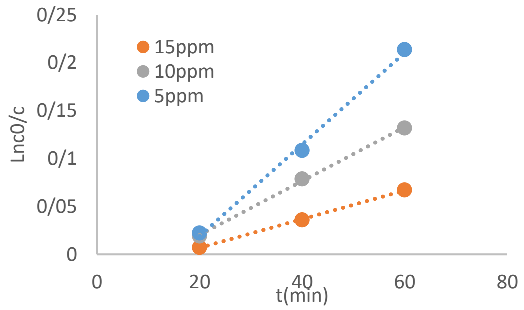


Figure 6. First-order kinetic model for congo red azo dye degradation at different concentrations, irradiated with a UV lamp in the presence of 9-layer mesoporous titanium dioxide film

Table 2. Adsorption rate constants of Congo red dye with different concentrations in the presence of a 9-layer mesoporous TiO_2 film (K)

k	Initial concentration
0.0048	5
0.0028	10
0.0015	15

Based on experiments performed on dye degradation at different acidic pHs (HCl), it was concluded that the higher the amount of acid added to the dye, the more dye degradation occurs.

The kinetics of dye degradation of 5 ppm Congo red in aqueous solutions of varying acidity were investigated under ultraviolet radiation in the presence of a 9-layer titanium dioxide film on a glass layer. As can be seen, the rate of dye degradation is higher in more acidic solutions.

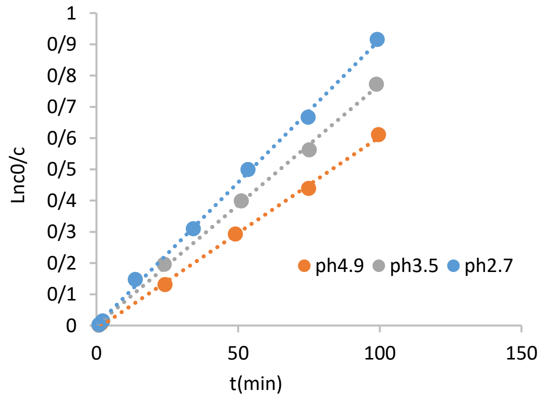


Figure 7. First-order kinetic model of dye degradation with a concentration of 5 ppm in aqueous solutions with different acidity, irradiated with ultraviolet radiation in the presence of 9-layer titanium dioxide film on a glass layer.

Table 3. The Effect of pH on the rate constants of congo red degradation with an initial concentration of 5 ppm using 9-layer TiO_2 films on a glass substrate (K)

K(min^{-1})	pH
0.0091	2.7
0.0077	3.5
0.0062	4.9

Furthermore, the performance of photocatalysts during repeated use is of great importance. For this purpose, the percentage of dye degradation in the presence of the 9-layer titania film was investigated in five experiments under identical conditions. The results are presented in Figure 8. As can be seen, with repeated use of the photocatalyst, dye degradation still occurs and no significant decrease in performance is observed after each reuse.

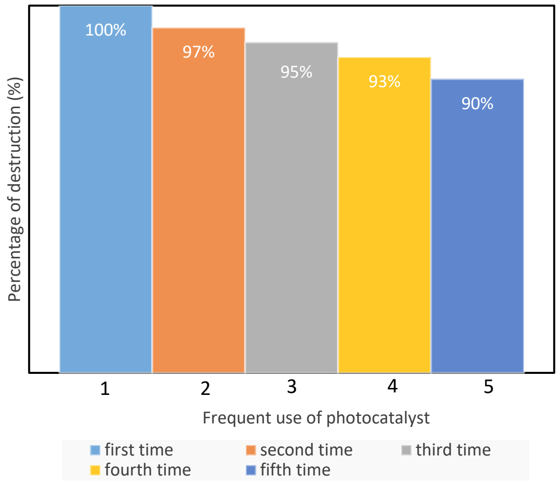


Figure 8. Evaluation of reusability (%) of 9-layer titanium dioxide mesoporous film for Congo red degradation.

The photovoltaic performance of a perovskite solar cell containing a 9-layer TiO_2 templated film with self-cleaning and UV-blocking properties is evaluated through current-voltage measurements. The current-voltage analysis is the most basic analysis of a solar cell, in which factors such as solar cell efficiency, open circuit voltage (Voc), short circuit current density (Jsc), and fill factor are determined. These parameters must reach a certain level of acceptability to state that a solar cell exhibits ideal photovoltaic behavior confidently. The J-V curve of the perovskite solar cell is presented in Figure 9, and the photovoltaic parameters measured under light irradiation are presented in Table 4. As can be seen, the results show that the perovskite solar cells prepared in this study exhibit good photovoltaic behavior and therefore perform well in converting sunlight into electricity. The slight decrease in photocurrent density in the presence of the titania UV blocking layer can be due to the higher refractive index of TiO_2 compared to glass.

Table 4. Photovoltaic performance of perovskite solar cells

Sample	V_{OC} (V)	J_{SC} (mA)	FF	η (%)
Without UV block	1	21.17	0.69	**14.58**
With UV block	1	20.40	0.68	**13.82**

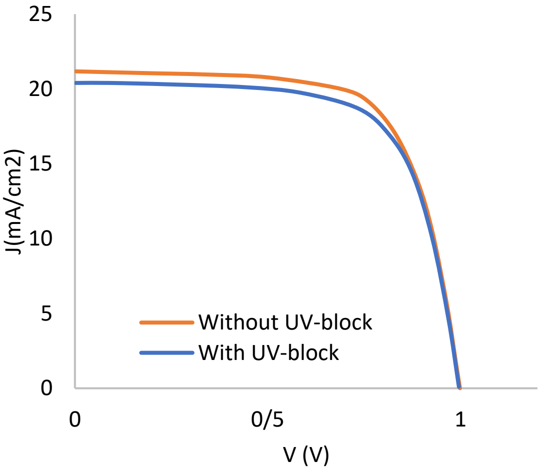


Figure 8. Current-voltage curves of the carbon-based perovskite solar cells with and without TiO_2 UV-blocking layer

Moreover, stability tests were conducted to investigate the stability of the prepared perovskite solar cell in the presence of a mesoporous TiO_2 UV-blocking layer. According to the results, the perovskite solar cell without a TiO_2 layer completely lost its stability in the presence of UV light after 20 days, while the carbon-based perovskite solar cell in the presence of a TiO_2 UV-blocking layer maintained its stability even after 30 days.

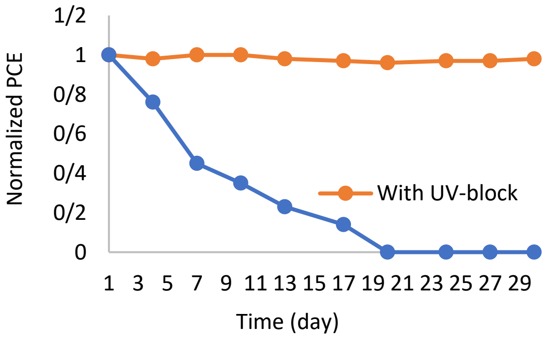


Figure 9. The stability test of carbon-based perovskite solar cells in the presence of UV light for 30 days.

In Table 5, a comparison of the stability tests and outcomes from the provided studies focuses on strategies to enhance stability through FTO/glass-side modifications or interfacial engineering. According to the comparison table, the carbon-based perovskite solar cell in this work, which was surrounded by a TiO2 UV-blocking layer, maintained its stability even after 30 days, more than another research.

Table 5. Stability comparison of perovskite solar cells device.

Modification	Test Conditions	Key Stability Outcomes	Ref
AZO/ZnO coatings on the glass side of FTO	100 h UV light ($50 mW/cm^2$) over 11 days	- AZO-coated: 40% initial efficiency retained - ZnO-coated: 30% retained	[7]
Amorphous TiO_2 (ALD) on Spiro-OMeTAD	Continuous AM 1.5G light + 0.85 V bias (120 h)	- TiO_2-coated: 96.5% PCE retained - Control: 20% loss	[23]
MgF_2 anti-reflective film on FTO glass	300 h illumination (AM 1.5G)	- MgF_2-coated: >70% PCE retained - Uncoated: Faster degradation	[24]
Mesoporous TiO_2 coatings on the glass side of FTO	100 h UV light ($50 mW/cm^2$) over 30 days	- Mesoporous TiO_2 -coated: 90% initial efficiency retained - Without coating: 10% retained	This work

Conclusions

In this study, we successfully developed a mesoporous TiO_2 templated film with dual functionality as a UV absorber and self-cleaning material for perovskite solar cells (PSCs). The film was fabricated using a dip-coating method from a solution containing P123 copolymer templates, resulting in a transparent mesoporous structure. The self-cleaning properties of the TiO_2 film were systematically evaluated through the photocatalytic degradation of Congo red dye, demonstrating its effectiveness in breaking down organic pollutants under UV light. Optimization of degradation conditions, including pH, dye concentration, and film thickness, revealed that the film exhibits superior photocatalytic activity under specific parameters, making it suitable for practical applications.

The integration of this TiO_2 film with carbon-based perovskite solar cells significantly enhanced their stability. Although the presence of this TiO_2 templated layer slightly reduced the PSC performance, on the other hand, in addition to creating self-cleaning properties, it also significantly increased its stability against UV light. Overall, this work highlights the potential of multifunctional TiO_2 films in addressing the stability issues of perovskite solar cells. The proposed approach offers a promising pathway for developing more durable and efficient PSC devices by combining UV protection and self-cleaning properties.

Conflict of Interest

There are no conflicts to declare.

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