Low-temperature Treated Anatase TiO$_2$ Nanophotonic-structured Contact Design for Efficient Triple-cation Perovskite Solar Cells

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Abstract

We report on the preparation and optimization of low temperature (<200°C) processed TiO₂ film as an electron transport layer (ETL) for high-performance perovskite solar cells (PSCs) compatible with flexible substrates. A high-quality ETL is spin-coated from hydrothermal synthesized single-phase crystalline anatase TiO₂ nanoparticles (NPs) with an average diameter of 6~10 nm. The surface of the high crystallite TiO₂ NPs reveals a tendency toward interparticle necking, facilitating compact scaffolds, resulting in PSCs with high power conversion efficiencies (PCEs). The influence of low and high temperature treated TiO₂ ETL on the device performance is studied. The best planar device fabricated in superstrate configuration (sup-C) exhibits a PCE of 17.1% with a $J_{SC}$ of 20.3 mA/cm². The PCE can be increased by ~25%, up to 23%, by moving from planar architecture in sup-C to the textured solar cell in substrate configuration (sub-C). The PSC covered with a nanophotonic-structured front contact allows gaining 8% and 15% on $V_{OC}$ and $J_{SC}$, respectively, where 2/3 of $J_{SC}$ gain is attributed to improved light incoupling, while the remaining 1/3 is due to increased diffraction at long wavelengths. The optical and electrical characteristics of the devices are investigated by 3D finite-domain time-domain (FDTD) and finite element method.
Keywords: Perovskite solar cells, Anatase TiO$_2$ NPs, Low-temperature process, 3D electromagnetic simulations

1. Introduction

In just a few years, perovskite solar cells (PSCs) have shown tremendous potential for next-generation photovoltaics due to outstanding improvements in power conversion efficiency (PCE). While PSCs with PCE of ~3.8% were first reported by Miyasaka and co-workers in 2009, intensive research efforts have successfully resulted in PSCs with a record PCE of 25.5% in the laboratory [1–3]. Perovskites offer excellent optoelectronic properties, such as high absorption, long carrier diffusion length, bandgap tunability, and low-cost solution-processed deposition [4–8], enabling perovskites further to realize efficient image sensors [9,10]. Generally, most high-efficiency perovskite photovoltaic device follows a sandwich structure with a perovskite layer placed between a metal oxide (MO) electron transport layer (ETL) and an organic hole transport layer (HTL) [6,11–14]. High-quality perovskite materials are usually deposited on the ETLs; hence, electron transport materials (ETMs) or hole blocking materials (HBMs) play a fundamental role in carrier extraction from PSCs, and also in eliminating electrical shunts to obtain high PCEs [7,8,15,16]. Besides, ETL is most commonly used as a front contact to realize perovskite-based high-efficiency tandem devices. The front contact of PSCs needs to be efficient so that it can fulfill several basic requirements, such as high conductivity, high transparency, and suitable electrical properties [17–20]. The titanium oxide (TiO$_2$) can fulfill all major front contact
needs. Hence, despite efforts to develop new materials, TiO$_2$ is still widely recognized as the preferred ETL for PSCs because of its environmentally friendly nature, tunable electronic property, cost-effectiveness, and conduction band (CB) matching that of perovskite, which makes it highly capable of electron injection and collection [8,21–24]. The deposition process has to be optimized so that high-quality perovskite can be formed on the ETL.

A compact ETL is a prerequisite to achieving high efficiency in a typical PSC structure (either planar heterojunction or mesoporous scaffold configuration) [11,25]. This is because of the tradeoff between ETL thickness and efficient carrier extraction, while a thin ETL allows fast electron extraction from the perovskite layer by blocking the recombination of electrons at the boundary with the perovskite layer [11,16,26,27]. A thick ETL can minimize short-circuits [22]. To date, few works have demonstrated PSCs without ETLs; nevertheless, the resulting device performance so far has been poor compared to that of PSCs that include ETLs [28,29]. Most significantly, high-performance PSCs use compact layer (CL) TiO$_2$ ETL that involves a high-temperature (>450 °C) sintering process, which increases fabrication costs and severely restricts their future applications in flexible, wearable, and low weight solar cells [8,11,26,30].

Hence, the preparation of low-temperature processed high-quality (uniform, pinhole-free, and compact) ETL is a prerequisite for the commercial deployment of PSCs. So far, very few works have been proven to realize low-temperature (<200 °C) treated compact ETL for the fabrication of PSCs [27,31–33]. Excellent prospects for using anatase TiO$_2$ NPs as a compact ETL in PSCs with high efficiencies have been previously confirmed. For example, improved PSC performance, PCE of 15.6%, has been demonstrated by Wang et al., who added graphene nanoflakes and anatase-TiO$_2$ NPs as ETL prepared at 150 °C [32].
In this study, high potential single-crystalline TiO$_2$ nanoparticles (NPs) were synthesized by the one-step hydrothermal method with a water-soluble titanium complex as a titanium source at low temperatures (<200 °C). The water-soluble titanium complexes contain less carbon content per titanium atom, making them very suitable as environment-friendly functional materials that can be considered potential ETL for PSC fabrications [34]. Herein, planar PSCs in the superstrate configuration have been investigated while TiO$_2$ NPs ETL was treated at low and high temperatures. The study further aims to design a nanophotonic solar cell that can deliver high PCE with optoelectronic properties. An advanced three-dimensional (3D) multiphysics approach that combines finite-difference time-domain (FDTD) and finite element method (FEM) along with the particle swarm optimization (PSO) algorithm has been developed and tested against fabricated planar PSCs to study optics, optimization, electrical characteristics. The adapted numerical approach provides an excellent agreement between experimental results and outcomes from simulations. A detailed description of the high-efficiency device design and optimization process is provided in the manuscript.

2. Results and Discussions

2.1 Properties of High-quality Anatase TiO$_2$ Nanoparticles and Perovskite Films

Pure phase, non-toxic, and environmental route single-crystalline anatase TiO$_2$ NPs were synthesized by a one-step hydrothermal process using a water-soluble titanium complex as a titanium source, as described by Tomita et al. [34]. Besides the environmental advantages of this water-soluble synthesis, these titanium complexes are successful reagents to lower the carbon content per titanium atom in titanium-containing functional materials. Figure 1(a-d) shows the scanning transmission
electron microscopy (STEM), Z-contrast, high-resolution transmission electron microscopy (HR-TEM), and corresponding selected area electron diffraction (SAED) pattern of the anatase TiO$_2$ nanoparticles, respectively. The TEM image reveals the spherical morphology of anatase TiO$_2$ NPs with particle sizes ranging from 6 to 10 nm in diameter (Figure 1(c)). The SAED pattern shows the characteristic (101) spacing, ensued in anatase phase only (Figure 1(d)), which is in agreement with the XRD. The surface of highly crystalline anatase TiO$_2$ facilitates interparticle necking and the formation of the compact scaffold. The spherical anatase TiO$_2$ forms denser scaffolds and offers multiple advantages, including efficient charge extraction, superior electron pathways, and, because of this dense packing, an exceptional hole-blocking effect [33]. Figure 1(e) shows the XRD patterns of anatase TiO$_2$ NPs with major diffraction peaks at 2θ of 25.4°, 38.28°, 47.46°, and 54.26° that correspond, respectively, to the (101), (004), (200), and (105) crystal planes of the tetragonal phase anatase TiO$_2$ (ICSD No. 9852). The mean crystallite size is 6.79 nm, as estimated by Scherrer’s formula ($D = \frac{K\lambda}{\beta\cos\theta}$), where $D$ is the crystallite size. $K$ is a dimensionless shape factor (0.94 for the spherical crystallite). $\lambda$ is the x-ray wavelength (0.15418 nm). $\beta$ and $\theta$ are line broadening at FWHM in radians and the Bragg angle, respectively. A detailed calculation method is provided in the literature [35]. The measured value is consistent with the particle size in the TEM image, indicating that each particle is composed of a single crystal. Therefore, the interior of the nanoparticles does not contain grain boundaries that can be recombination sites, which is suitable for charge transport. In addition to the XRD pattern, the Raman spectra in Figure 1(f) did not show any other phases such as rutile type [36], confirming that the synthesized sample is a highly pure anatase type nanoparticle, suggesting that it had a minimized number
of surface defect traps. This may lead to enhanced charge transport at the TiO$_2$/perovskite interface.

Figure 1. (a) Scanning transmission electron microscopy (STEM) image, (b) Z-contrast image, (c) high-resolution transmission electron microscopy (HR-TEM) image, (d) corresponding selected area electron diffraction (SAED) pattern, (e) XRD pattern, and (f) Raman spectra of anatase TiO$_2$ NPs.

The Brunauer–Emmett–Teller (BET) analysis is a well-known method for determining the surface area of nanoparticles. The Brunauer–Emmett–Teller (BET) analysis shown in Figure S1 confirms that the anatase TiO$_2$ NPs had a large specific surface.
The surface area of 170.2 m$^2$g$^{-1}$. Using this specific surface area, the BET diameter was determined to be 8.3 nm assuming the uniform size, spherical shape, which was consistent with the particle size from the TEM image (Figure 1(c)) and crystallite size calculated from XRD (Figure 1(e)). Further information about the BET calculation is available in the Supporting Information and literature [37,38].

The optical properties of deposited TiO$_2$ NPs films can be further understood by UV-vis measurement. Figure S2 in the Supporting Information shows the absorbance and transmission of the fabricated TiO$_2$ NPs films on FTO substrates treated at 200 °C and 450 °C, respectively. Both films exhibit almost identical characteristics without showing a significant discrepancy. The top-view SEM images for the five-coating cycle anatase TiO$_2$ NPs films treated at 200 °C and 450 °C are shown in Figure 2(a) and Figure S3(d) (Supporting Information), respectively, confirming that both films have similar surface morphologies that are uniform, dense, and pinhole-free. This is further confirmed by atomic force microscopy (AFM), as presented in the Supporting Information (Figures S3(e) and S3(f)), where the root means square (RMS) roughness is calculated to be 3.5 and 4.0 nm, respectively, for the films treated at 200 °C and 450 °C. The above data shows excellent thin-film morphology with low roughness. The smooth, pinhole-free, and dense scaffolds efficiently block direct electrical contact between the FTO and perovskite active layers. A large number of anatase domains are expected to provide efficient electron transfer characteristics, while the ability to independently control the film thickness without affecting the morphology or crystallographic characteristics enables a route to optimize efficient electron extraction and hole blocking in the fabricated PSCs. Figures 2(b) display the cross-section SEM image of the resulting anatase TiO$_2$ NPs film on FTO substrates for the optimum five-coating cycles, where one-, three-, and seven-coating cycles'
cross-sectional SEM images are provided in the Supporting Information (Figures S3(a-c)), which confirm the formation of uniform and dense scaffolds with increasing thickness as the number of cycles increases.

**Figure 2.** (a) Top-view SEM and (b) cross-section images of anatase TiO$_2$ NPs processed at 200 °C for the optimum five-coating cycle. The inset in (a) shows the top-surface view of anatase TiO$_2$ NPs processed at 200 °C with a different scale. (c) Top-view SEM micrographs of perovskite film deposited on the TiO$_2$ NPs/FTO substrate treated at (c) 200 °C and (d) 450 °C. (e) The XRD patterns of perovskite films fabricated with the TiO$_2$ NPs films treated at 200 °C and 450 °C. (f) PL spectra of FTO/CsFAMA, FTO/TiO$_2$ NPs (treated at 200 °C)/CsFAMA, and FTO/TiO$_2$ NPs (treated at 450 °C)/CsFAMA films.
Cesium-Formamidinium-Methylammonium (termed CsFAMA) triple cation-based perovskite films were fabricated on TiO\textsubscript{2} NPs films prepared at 200 °C and 450 °C on FTO substrates. Previously, we reported that perovskite films grown on TiO\textsubscript{2} films (treated at low- and high-temperature) present similar PL quenching, which implied efficient electron transfer from the perovskite film to the ETLs [39]. The perovskite film on FTO-TiO\textsubscript{2} NPs (treated at 200 °C) ETL exhibits more significant quenching than perovskite film on FTO-TiO\textsubscript{2} NPs (treated at 450 °C), revealing efficient charge transfer at the interface between the photoexcited perovskite and the TiO\textsubscript{2} NPs. Figure 2(c, d) shows the top-view SEM image of the perovskite film fabricated with the anatase TiO\textsubscript{2} NPs treated at 200 °C and 450 °C, respectively. It is observed that low-temperature treated samples contain slightly large crystal grains with uniform and flat surface morphology compared with the anatase TiO\textsubscript{2} NPs treated at 450 °C. This is consistent with good interparticle adhesion across the interface between perovskite and single-crystalline anatase TiO\textsubscript{2} NPs. Fewer nucleation sites are produced that further facilitate the perovskite nucleation because of the inferior nucleation energy boundary at the interface [33,40]. Subsequently, perovskite growth leads to a smooth interface containing fewer grain boundaries with larger crystal domains, a more continuous and uniform morphology, and enhanced crystallinity. Primary nucleation is dominated by the resultant pure-phase compact anatase TiO\textsubscript{2} NPs. Previously, Miyasaka and co-workers revealed that the growth of the perovskite layer depends sensitively on the quality and chemistry states of the substrates [41]. Large anatase single crystals present superior mobility created by thermal excitement of shallow donor states as described by Forro et al. [42]. Single-crystalline anatase TiO\textsubscript{2} NPs provide additional benefits compared to the compact TiO\textsubscript{2} layer due to the large interface area for efficient
electron collection and transport, which further balances the charge carriers. In our case, the relatively large perovskite crystals with fewer grain boundaries result from the single-crystalline anatase TiO$_2$ NPs. The resulting XRD spectra in Figure 2(e), showing no significant difference for both TiO$_2$ NPs annealing conditions, present diffraction peaks at $2\theta = 14.14^\circ$, $20.0^\circ$, $28.40^\circ$, $31.91^\circ$, $40.74^\circ$, and $52.9^\circ$ that can be assigned, respectively, to the (110), (112), (220), (310), (224), and (440) planes of the key phases of perovskite [33]. Significantly, no peak from PbI$_2$ at $12.6^\circ$ could be detected in either sample indicating the complete transformation of PbI$_2$. The full width at half maximum (FWHM) of the (110) diffraction peak for perovskite films fabricated with the TiO$_2$ NPs films treated at 200 °C and 450 °C were 0.10 and 0.11, respectively. Together, these results suggest a very similar crystallinity for the perovskite films irrespective of the TiO$_2$ NPs film processing temperature. To gain insight into the recombination behaviors of charge carriers in the fabricated devices, the PL emission spectra of FTO / CsFAMA, FTO / TiO$_2$ NPs (treated at 200 °C) / CsFAMA, and FTO / TiO$_2$ NPs (treated at 450 °C) / CsFAMA films were measured, and the findings are shown in Figure 2(f). Notably, PL quenching implies efficient electron transfer from the perovskite to ETLs. These advantages are expected to realize the PSCs with high performance.

2.2 Experimental Verification of Perovskite Solar Cells

A schematic of the fabricated device design with the corresponding energy level diagram is depicted in Figure S4 in the Supporting Information. A cross-sectional FESEM image of the fabricated planar PSC with the anatase TiO$_2$ NPs film treated at 200 °C is shown in Figure 3(a). The perovskite absorber is placed between TiO$_2$ NPs ETL and Spiro-OMeTAD HTL, allowing efficient charge carrier extractions. The
fabricated perovskite film is 350 nm thick, whereas the HTL and Au metal contact thickness is 250 nm and 100 nm, respectively. To examine the effects of anatase TiO$_2$ layer thickness on the PSC performance, a series of devices were fabricated while varying the number of TiO$_2$ coating cycles; see Figure 2 and Figure S3.

![Cross-sectional FE-SEM image of the fabricated champion planar PSC with the low temperature treated TiO$_2$ ETL](image)

**Figure 3.** (a) The cross-sectional FE-SEM image of the fabricated champion planar PSC with the low temperature treated TiO$_2$ ETL. (b) A comparison of J-V characteristics between experiment (forward scan (FS) and reverse scan (RS)) and simulations for the best-performing planar PSCs. Inset Figure (b) shows the extracted photovoltaic parameters, and the TiO$_2$ NPs have an optimized thickness of 80 nm. (c) Steady-state stabilized efficiency was measured for the five-cycle coating of TiO$_2$ NPs with the low temperature (200 °C)-based device. (d) A comparison of QEs between experiments and simulations for the best-performing planar PSCs.

The five-coating cycle (80 nm TiO$_2$ film) shows the best photovoltaic performance, exhibiting comparatively lower hysteresis. The J-V characteristic of the champion fabricated planar PSC with the reverse scan (RS) for the anatase TiO$_2$ NPs film treated at 200 °C is shown in Figure 3(b). The J-V characteristics of fabricated PSCs in the
forward scan (FS) and RS directions with the low temperature treated TiO\(_2\) for different coating cycles are provided in the Supporting Information (Figure S5). As a reference, the J-V characteristics of the fabricated PSCs in the FS and RS directions for high temperature (450 °C) treated TiO\(_2\) with 5-coating cycle TiO\(_2\) NPs films are shown in Figure S6. The photovoltaic parameters of corresponding champion devices in the case of 200 °C and 450 °C are provided in Table 1. Error bars indicate plus or minus one standard deviation from the mean.

### Table 1. Comparing photovoltaic performance parameters of investigated PSCs extracted from J-V characteristics, as shown in Figure 3(b) and S6.

<table>
<thead>
<tr>
<th>Anatase TiO(_2) NPs</th>
<th>Scan Direction</th>
<th>(J_{SC}) (mA/cm(^2))</th>
<th>(V_{OC}) (V)</th>
<th>FF (%)</th>
<th>PCE (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>200 °C</strong></td>
<td>Forward</td>
<td>20.08</td>
<td>1.02</td>
<td>73</td>
<td>15.0</td>
</tr>
<tr>
<td></td>
<td>Reverse</td>
<td>20.33</td>
<td>1.09</td>
<td>79</td>
<td>17.10</td>
</tr>
<tr>
<td></td>
<td>Average±S D</td>
<td>18.62±1.22</td>
<td>1.05±0.05</td>
<td>0.68±0.09</td>
<td>13.52±2.65</td>
</tr>
<tr>
<td><strong>450 °C</strong></td>
<td>Forward</td>
<td>19.81</td>
<td>1.07</td>
<td>57</td>
<td>12.2</td>
</tr>
<tr>
<td></td>
<td>Reverse</td>
<td>19.45</td>
<td>1.08</td>
<td>72</td>
<td>15.06</td>
</tr>
<tr>
<td></td>
<td>Average±S D</td>
<td>19.02±2.08</td>
<td>1.02±0.08</td>
<td>0.61±0.06</td>
<td>11.97±2.97</td>
</tr>
</tbody>
</table>

All J-V characteristics were analyzed under one sun condition, AM 1.5G (100 mW/cm\(^2\)), and all devices were measured in air with humidity fluctuating from 30% to 40% at 20 °C without encapsulation. In the five-coating cycle, the resultant TiO\(_2\) NPs based perovskite film contains a more apparent grain size with refined grains and fewer disorganized lattices, which potentially solves unbalanced ion aggregation close to the grain boundaries and ensures slower electrode polarization. As a result, a more substantial electron extraction efficiency is achieved, facilitating the reduction of charge carrier recombination. Thus, the five-coating cycle TiO\(_2\) NPs layer-based PSCs performed more efficiently than the TiO\(_2\) NPs-based counterparts, indicating the
presence of well-organized charge transfer at the TiO$_2$/perovskite interface. As shown in Table 1, PSCs with the low-temperature processed TiO$_2$ NPs exhibit superior performance compared to the high-temperature processed TiO$_2$ NPs, which allows further utilizing such ETLs for several promising efficient device applications. To confirm stabilized efficiency of the resultant hysteretic cells, we performed steady-state photocurrent measurement at the maximum power point for the device with TiO$_2$ NPs film treated at low-temperature (200 °C). Figure 3(c) exhibits the stabilized efficiency under 1 sun (AM 1.5G) irradiation. The resultant device shows a stabilized efficiency of 17% (up to 150 s), which is very close to the efficiency estimated from J-V measurement in reverse scan (RS) (Figure 3(b)). The stabilized efficiency evidences the impact of larger crystal domains with fewer grain boundaries (i.e., gaps and traps) on the PSC efficiency.

The incident photons to electron conversion were also measured to verify the performance of the solar cell. Figure 3(d) shows the quantum efficiency (QE) of optimum 80 nm thick TiO$_2$ based PSC, where QEs of other devices with the coating cycle variation is presented in Figure S7. The five-coating cycle case demonstrates higher QE throughout the complete spectrum (300 nm to 800 nm) with 80-85% peak intensity, as compared to other cases. Such a broader photon absorption further allows collecting more electrons effectively at the perovskite/TiO$_2$ edge due to the reduced limit of the interfacial energy barrier. The QE of the PSC with the TiO$_2$ film treated at 450 °C is provided in the Supporting Information (Figure S8) for reference. The QE of the champion PSC with the anatase TiO$_2$ NPs film treated at 200 °C contributed to a considerably high short-circuit current density ($J_{sc}$) of 20.33 mA/cm$^2$, open-circuit voltage ($V_{oc}$) of 1.07 V, fill factor (FF) of 79%, and PCE of 17.10% in the
Moreover, the PSC reproducibility with the TiO$_2$ ETL treated at 200 °C and 450 °C was studied by fabricating 22 and 13 individual devices, respectively, where photovoltaic parameters ($J_{SC}$, $V_{OC}$, FF, and PCE) of each case is plotted in Figure S9 in the Supporting Information. Overall, the TiO$_2$ ETL treated at 200 °C devices show a larger average PCE of 13.52 ± 2.65% with better reproducibility than TiO$_2$ ETL treated at 450 °C devices (11.97 ± 2.65%). A comparison of the photovoltaic performance between our fabricated device and relevant best-reported devices in the literature is provided in Table S1 (Supporting Information). The basic planar solar cell structure’s device performance is also much higher than our recently published works in quality journals.[8,11,21,39] Also, while the PSC with the high-temperature treated TiO$_2$ NPs film show lower hysteresis, its performance, $J_{sc}$ of 19.45 mA/cm$^2$, $V_{OC}$ of 1.08, FF of 72%, and PCE of 15.06% in the RS direction, lower than for the low-temperature treated TiO$_2$ case. Nevertheless, we emphasize that while it might be possible to enhance the performance of the device integrated with the high-temperature treated TiO$_2$ NPs to, for example, facilitate efficient electron flow from ETL to perovskite, this is not the scope of the current study. Instead, in this work, we focus exclusively on the prospects of designing high-efficiency PSC using solely low-temperature treated TiO$_2$ NPs ETL. In this case, our results demonstrate that thermal annealing at a high temperature is not always necessary to achieve high-efficiency PSCs with greater device stability. To confirm this, we further performed light soaking and moisture stability measurements to evaluate the reliability of the resultant PSC. The light soaking test was carried out by continuous exposition of non-encapsulated devices to 1 sun illumination for ~4800 seconds in humid air at the RH ranging from 30% to 40%. The resultant device retained over 70% of its initial efficiency after the light-soaking
investigation (Figure S10). This stabilizes device PCE due to the improved perovskite film quality. The resultant device showed no noticeable degradation in $J_{SC}$ and $V_{OC}$ even after continuous exposure of light over 4800 secs. Moreover, the $J$–$V$ curve of the device was assessed before and after dark storage at ambient atmosphere (without sealing or encapsulation). The averaged photovoltaic parameters (normalized $J_{SC}$, $V_{OC}$, FF, and PCE) versus time (secs) are presented in Figure S10(a-d). In the following step, the QE and J-V characteristic of the solar cell is investigated. A coupled optical-electrical 3D numerical simulation approach was adopted to study the planar PSCs in superstrate configuration. A 3D method is used for 3D textured solar cells to be investigated in subsequent sections. The generation rate calculated from the optical simulation is used as one of the inputs for the FEM simulation of the J-V characteristics. Details on the simulation methods used are provided in Section 4.5. We compared the experimentally obtained results with the outcome of the optoelectrical coupled numerical simulations [7,12], so that the realistic photovoltaic performance can be achieved from the future investigation. A reasonable agreement between the experimentally measured QE and $J_{SC}$ and the simulated curves is reached, as shown in Figures 3(b) and 3(d). The photovoltaic parameters of the simulated planar device in superstrate configuration are provided in Table 2. Interference fringes are more pronounced for the simulated QE because the solar cell interfaces are assumed to be completely flat, while some roughness characterizes real interfaces. The simulation of the experimental results allows for determining an optical and electrical model of the solar cells. The optical wave phenomena happening in the flat device in superstrate configuration can be further understood by simulated power densities and electric field distributions provided in the Supporting Information (Figure S11).
2.3 Nanophotonic Front Contact Design for Efficient Perovskite Solar Cells

The low-temperature treated anatase TiO$_2$ NPs film has a substantial interest for flexible tandem solar cell (TSC) applications [43,44]. Furthermore, the low-temperature process allows for the formation of TiO$_2$ films on top of perovskite films. Hence, it allows for the realization of solar cells in a substrate configuration [45,46]. Solar cells in substrate configuration have the potential to reach higher PCE due to an optical design that allows for improved coupling of light into the solar cell material.

Although the fabricated basic planar PSC is highly efficient, it does not allow, in most cases, the monolithic deposition of top cells on bottom cells.[19,46] In the following, the front contact of a PSC in substrate configuration is designed. The front contact must fulfill several functions [15]. Most authors propose a front contact consisting of a multilayer system to fulfill the different requirements [15,47]. The first layer directly prepared on the perovskite layer has to prevent the damage of the underlying layer; secondly, the layer has to exhibit a suitable work function to allow for an efficient charge extraction and a high $V_{OC}$. Furthermore, the front contact should provide high lateral conductivity to allow for a high FF of the solar cell. Thirdly, the front contact's absorption loss should be low, and the front contact should couple the light into the solar cell and potentially scatter or diffract the incident light. Nevertheless, the utilization of such a front contact is not possible in the presently fabricated device structures. So far, we have studied only the PSC in the superstrate configuration. From now on, we will focus on the substrate configured PSCs, which is opposite to the superstrate configured device. A schematic cross-section of the planar PSC integrated with a multilayer front contact is displayed in Figure 4(a), and corresponding layer information is provided in Table S2 in the Supporting Information. The multilayer front
contact consists of TiO$_2$ and TCO layers, where the TCO is placed between TiO$_2$ NPs and optically lossless TiO$_2$ layer [24]. The synthesized anatase TiO$_2$ NPs acts as a potential ETL to improve electron extraction and transportation from the perovskite absorber, while thick optically lossless TiO$_2$ helps to improve light incoupling. A doped ZnO with a conductivity of ~1000 S/cm was considered as a TCO in the current study.

**Figure 4.** (a) A schematic cross-section of the multilayer front contact-based flat PSC in substrate configuration. Corresponding power density profile for an incident wavelength of (b) 400 nm, (c) 550 nm, and (d) 700 nm, respectively. (e) A schematic cross-section of the substrate configured front textured PSC. Corresponding power density profile for an incident wavelength of (f) 400 nm, (g) 550 nm, and (h) 700 nm, respectively. The texture has a period of 600 nm and a height of 600 nm.

The TCO should have a thickness range of 50–400 nm otherwise, the device's efficiency will be restricted due to poor optical and electrical properties. More details
about the optoelectronic performance and TCO effects in PSCs are provided in previously published works [15,18]. It is noted that other TCO materials (e.g., ITO, FTO, IZO) can also be used in the proposed device design. Simulated power densities of the device under the monochromatic wavelength of 400 nm, 550 nm, and 700 nm are presented in Figures 4(b-d). The optical wave propagation within the substrate configured solar cell is comparable with that of the superstrate configured solar cell, as understood by Figure S11. At 400 nm wavelength, most incident photons are absorbed in the vicinity of the TiO$_2$ ETL and perovskite layer, where all photons below 400 nm are absorbed by the front contact that does not contribute to the QE or $J_{SC}$ and is considered as a parasitic loss. In addition to that, a fraction of the incident light propagates to the bulk of the perovskite absorber. An increase of the wavelength (550 nm), leads to the enhancement of the absorption only in the perovskite layer, as shown in Figure 4(c). At 700 nm, most photon absorption occurs in the active layer; however, a slight absorption loss occurs in the back contact at the HTL/metal interface. After 800 nm wavelength, no absorption is recorded in the active layer due to the optoelectronic properties of the perovskite materials. Such absorption mechanisms are confirmed by the QE plots, as shown in Figure 5(b), where a comparable absorption (almost equal $J_{SC}$) is found for both flat solar cells. The optical wave propagation can be further appropriately understood by the electric field distribution on the flat device, which is shown in Figure S12(a-d) in the Supporting Information. Nevertheless, substrate configured flat PSC pronounces higher fringes than the superstrate configured flat device, mainly due to thicker TiO$_2$ and air/metal-oxide interfaces. Similar patterns are also found in their corresponding reflections, shown in Figure 5(d). In the investigated PSC, multilayer metal oxides are used to form a front contact, which has a comparable refractive index (~2.5) with the perovskite material.
system, leading to reduce the reflection as a stack of metal oxides and perovskite considered as a single block and act as the refractive index gradient [48]. However, the planar device yet exhibits considerable reflections due to the flat surface at the front, which restricts to absorption of adequate photons in the device.

**Figure 5.** (a) A graphical representation of square-based dome shape periodic nanostructures ($p$ and $h$ are the period and height of the dome). A comparison of (b) quantum efficiencies and (d) corresponding reflections between flat (superstrate and substrate configurations) and textured PSCs. (c) A comparison of $J-V$ characteristics between flat (superstrate and substrate) and textured PSCs. The texture has a height of 600 nm and a period of 600 nm.

Hence, it is essential to improve light incoupling and/or light trapping to realize efficient photon management. Efficient photon management allows reducing reflections and enhancing $J_{SC}$ as well as PCE. In this study, only the front contact is textured by using
square-based dome-shaped nanostructures that lead to achieving improved light incoupling and light trapping in the device. In the proposed design, the interface between ETL and perovskite is kept flat so that effective electrical properties cannot deteriorate. Figure 5(a) depicts a periodic arrangement of the nanostructures, where \( p \) and \( h \) define the period and height of the domes, respectively. The proposed PSC design covered with front nanostructures is shown in Figure 5(e). The designed textured PSC can significantly contribute to light incoupling by facilitating a broadband anti-reflection coating and further improving photon absorptions in the longer wavelength region [48–50]. The fabrication of such solar cells is simple and comparable to the planar device, where nanostructures could be prepared on the top of the planar device by industrially viable imprinting or stamping technique, or even some form of colloidal lithography method [51–54]. In the current study, the optimum dimension of the dome shape nanostructures is calculated by using Particle Swarm Optimization (PSO) algorithm embedded with the FDTD solver [24,55]. It has been found that the photovoltaic performance is only maximized while the texture has a height and period of 600 nm. Figure 5(b) demonstrates the QE of the optimized PSC along with the QEs of the flat reference devices in superstrate and substrate configurations. A substantial improvement of the QE of the optimized device has been observed compared to that of flat solar cells throughout the complete spectral range, where fringes of the QE are also considerably reduced. The improved light incoupling and light trapping facilitate the QE approaching the unity at the wavelength range of 600-650 nm. The improved QE leads to an increase in the \( J_{SC} \) of the optimized PSC. In contrast, the optimized device's reflection losses are significantly reduced compared to the flat device, as shown in Figure 5(d). It has been further noted that the QE is shifted to the visible-infrared region, which additionally lets attaining a stable device
Optics of the optimized PSC is also understood by its simulated power densities and electric field distributions. Figure 4(f-h) demonstrates the power densities of the optimized structure under the monochromatic wavelength of 400 nm, 550 nm, and 700 nm, where electric field distribution profiles are provided in the Supporting Information (Figure S(12(e-h)).

It is confirmed that the nanophotonic designed front contact contributes to the enhanced light incoupling and facilitates light trapping via diffraction of the incident light, leading to an improved PCE of the solar cell. The optical path length of the propagating wave is increased, which leads to attaining improved photon absorptions.

In the proposed device design, we considered the TiO$_2$ for preparing front nanostructures (Figure 4(e)); however, it is feasible to utilize other materials for the nanostructure. A comparison of QEs, reflections, and absorptions of textured solar cells in substrate configuration while using different materials to make nanostructure is provided in the Supporting Information (Figure S13) along with their schematic device structures. It has been found that the QE is comparatively higher in the case of TiO$_2$; nevertheless, the absorption is maximized for the ZnO case. It is investigated that the shifting of planar solar cells from superstrate architecture to substrate architecture enhances photovoltaic performance for a certain level; however, as compared to the optical properties, the electrical properties of solar cells are much improved. Consequently, the nanostructured solar cell in the substrate configuration further enhances the optical properties considerably, leading to enhanced PCE.

Figure 5(c) illustrates the comparison of J-V characteristics between flat reference devices (superstrate and substrate configurations) and optimized PSC, and corresponding extracted photovoltaic parameters are summarized in Table 2. As compared to the flat PSC in superstrate configuration, the $J_{SC}$ of the optimized PSC in
substrate configuration is improved by 13.5%, leading to the maximum $J_{SC}$ of 23.5 mA/cm$^2$ while the $V_{OC}$ is enhanced by 8%, up to 1.19 V. It has been noticed that the electrical parameter ($V_{OC}$ and FF) values of flat and textured solar cells in substrate configuration are almost the similar, which confirms the practical design of the proposed PSC. The PCE of the proposed textured solar cell is increased by 25-30% compared to that of reference simulated and fabricated best planar devices in superstrate configuration. The investigated PSC integrated with the nanophotonic front contact exhibits an estimated PCE of 23% with a $J_{SC}$ of 23.5 mA/cm$^2$, $V_{OC}$ of 1.186 V, FF of 83%. These results suggest that the investigated nanophotonic multilayer front contact design has the potential for realizing high-efficiency single-junction PSCs as well as perovskite-based TSCs [12,56].

Table 2 Comparing photovoltaic performance parameters of investigated PSCs extracted from J-V characteristics, as shown in Figure 5c.

<table>
<thead>
<tr>
<th>Device Structure</th>
<th>$J_{SC}$ (mA/cm$^2$)</th>
<th>$V_{OC}$ (V)</th>
<th>FF (%)</th>
<th>PCE (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flat (Superstrate)</td>
<td>20.3</td>
<td>1.1</td>
<td>81</td>
<td>18.0</td>
</tr>
<tr>
<td>Flat (Substrate)</td>
<td>21.1</td>
<td>1.19</td>
<td>83</td>
<td>20.8</td>
</tr>
<tr>
<td>Texture (Substrate)</td>
<td>23.5</td>
<td>1.186</td>
<td>83</td>
<td>~23</td>
</tr>
</tbody>
</table>

We now turn our attention to the feasibility of fabricating the proposed nanostructures with the required tolerance specifications. It is first noted that current nanofabrication technologies are highly precise; the proposed nanostructures can be fabricated viable soft imprinting technique, which has an excellent transfer process, desirable controllability, and reproducibility [52,57–59]. To study the performance of proposed PSCs under realistic conditions, we then perform computational simulations for several values of the main nanotexture parameters to provide an order of magnitude of the impact of differences between the design and practical demonstration. For this
purpose, we vary the D and H parameters of the nanotexture from 600 nm to 580 nm and investigated the device performance for single-texture and multi-structured front contacts. The results show that these changes in the nanotexture profile dimension do not significantly affect the solar cells' performance, as shown in Figure S14, Figure S15, and Table S3 in the Supporting Information. These results are consistent with published results for similar nanotextures, prepared via soft imprinting technique, showing that nanotexture variability does not significantly affect the electrical properties ($V_{OC}$ and FF) of the device [59]. We, therefore, conclude that small deviations in the actual fabricated patterns concerning the numerically optimized patterns, will not result in substantial differences in performance. Accordingly, we conclude that experientially validated numerical modeling, as presented in this work, provides a valuable and reliable strategy to advance PSCs’ through the current research and commercialization challenges [60,61]. In addition to that, the proposed synthesized novel single-crystalline anatase TiO$_2$ NPs treated at low temperatures are environmentally friendly due to the lower carbon content in titanium-containing functional materials. Furthermore, single-crystalline dense anatase TiO$_2$ NPs has more benefit than compact TiO$_2$ layer due to the large interface area for election injection and the following transport that further balances the charge carriers. The proposed device allows improved charge transport and extraction, leading further to realize flexible and high-efficiency tandem solar cells.

3. Summary

The prospect of utilizing low temperature (<200 °C) treated TiO$_2$ nanoparticles (NPs) as the potential ETL for high-efficiency perovskite solar cells (PSCs) has been
systematically investigated. TiO$_2$ NPs were prepared by the low-cost spin coating technique, allowing deposition on a wide range of flexible substrates (e.g., plastic, paper-based materials, metal foils, fabrics, etc.). We have optimized the ETL thickness for realizing highly efficient planar PSCs by changing the coating cycle during the fabrication of devices, solely used TiO$_2$ NPs as the ETL-based planar PSC demonstrated excellent photovoltaic performance with a PCE of 17.10%, $J_{SC}$ of 20.33 mA/cm$^2$, $V_{OC}$ of 1.07 V, and FF of 79%. We utilized 3D finite-difference time-domain (FDTD), and finite element method (FEM) based opto-electrical coupled simulations to investigate optics, optimizations, electrical characteristics of the solar cells. The outcome from numerical approaches nicely validates the experimental data that further optimize and realize high-efficiency PSCs. In the current study, we designed a multilayer nanophotonic front contact for the PSC with high efficiency by maintaining flat interfaces between perovskite and contacts, providing better electrical properties. The nanophotonic-structured front contact allows improving light incoupling and light trapping that contributes to the enhancement of $J_{SC}$ by 12~18% and PCE by 18~30%. The optimized PSC reveals an estimated PCE of 23%, with a $J_{SC}$ of 23.5 mA/cm$^2$, $V_{OC}$ of 1.186 V, FF of 83%. Hence, it is indicated that low-temperature treated TiO$_2$ NPs can be an ideal ETL candidate as a scaffold for high efficiency flexible solar cells. This study can further enable the fabrication of entirely low-temperature processed all perovskite tandem solar cells, contributing to the fabrication of low-cost, flexible solar modules in the future.

4. Methods

4.1 Materials
All materials were purchased from commercial suppliers and used as received. FTO-coated glass substrates were purchased from Asahi glass (Tokyo, Japan). The perovskite precursors consisted of methylammonium bromide (MABr), and formamidinium iodide (FAI), cesium iodide (CsI), lead bromide (PbBr₂), and lead iodide (PbI₂) were bought from TCI (Tokyo, Japan). N, N-dimethylformamide (DMF, purity 99.5%) and Dimethyl sulfoxide (DMSO, purity 99.5%) were purchased from Wako Pure Chemical (Osaka, Japan). Anatase TiO₂ NPs were synthesized according to the literature procedures.[34] Since then, the TiO₂ NPs were dispersed in distilled water and prepared colloidal suspension.

4.2 Characterization

Solar cell characteristics (current density versus voltage measurement) of all devices were measured with a scan rate of 0.05 V/s with forward-scan (FS; from -0.1V to 1.2V) and reverse-scan (RS; from 1.2V to -0.1V) with an active area of 0.09 cm² under 100 mW/cm² AM 1.5G simulated by a solar simulator using a Keithley 2401 digital source meter. The incident photon-to-electron conversion efficiency (IPCE) spectra of the device were measured using a monochromatic xenon arc light system (Bunkoukeiki, SMI-250JA). Whole devices were measured in air with humidity differing from 50% to 60% at 20 °C. The field emission scanning electron microscopy (FE-SEM; S-4800, Hitachi High-Tech, Tokyo, Japan) together with scanning transmission electron microscopy (STEM; HF-2200 TU, Hitachi Ltd., Tokyo, Japan) were used to analyze the surface morphology and NPs diameter size, respectively. Raman spectroscopy (STR150, AIRIX, Tokyo, Japan) using 50 mW-532 nm diode laser was used to measure further confirmed selectively synthesized AT TiO₂ NPs. The specific surface areas measuring device (BET; Bellsorp mini II, Microtrac BEL, Tokyo, Japan) were measured to know the particular surface area of AT TiO₂ NPs. The X-ray
diffraction (XRD) patterns of TiO$_2$ nanocrystals and perovskite films were measured using an X-ray diffractometer (D8 Discover, Bruker AXS Co. Ltd, Tokyo, Japan) with an X-ray tube (Cu Kα radiation, $\lambda = 1.5406$ Å).

4.3 Synthesis of Anatase TiO$_2$ Nanoparticles

Nanocrystalline anatase TiO$_2$ NPs were synthesized by hydrothermal conditions using a water-soluble titanium complex as a titanium source. Finely powdered titanium of 10 mmol was dissolved in the mixed solution of 30%-H$_2$O$_2$ aq. and 28%-NH$_3$ aq., then malic acid of 10 mmol was added here. After the solution was dried at 50 °C to remove excess H$_2$O$_2$ and NH$_3$, 0.5 M Ti-malate complex solution was prepared by adding distilled water. The Ti complex solution was heat-treated in an autoclave at 200 °C for 5 h. Formed TiO$_2$ particles were separated by centrifuge and washed with distilled water twice. TiO$_2$ NPs were dispersed in distilled water.

4.4 Device Fabrication

FTO-patterned glass substrates were sequentially cleaned in commercial detergent in water, deionized water, acetone, and ethanol in a sonication bath, each for 15 min. The cleaned FTO-coated substrates were then dried with a nitrogen stream and treated by UV-Ozone for 15 min. As the synthesized noncolloidal suspension of pure-phase anatase, TiO$_2$ NPs solution was spin-coated with varying cycle-coating onto FTO-substrates by spin-coating method at 2000 rpm for 30 s and annealed at 100 °C for 5 min, followed by annealing at a low-temperature (<200 °C) for 1 h and at a high-temperature (> 450 °C) for 30 min. This spin-coating and annealing process was cycled several times to control its thickness and minimize potential pinhole overlapping. Then, the triple cation-based precursor solution with Cs 5% was prepared by mixing with FA/MA in DMF and DMSO with the ratio (4:1) mixed solvents. The detailed solution making and spin coating procedure was reported elsewhere. The stock
solution of FAI (1 M), PbI$_2$ (1.1 M), MABr (0.2 M), and PbBr$_2$ (0.2 M) were mixed in anhydrous DMF: DMSO (4:1), and an appropriate volume of CsI solution was added from CsI stock solution (1.5 M) in DMSO to achieve Cs 5% solution. The perovskite solution was spin-coated in three steps: with a first step 0 rpm for 10 s, then in the second step at 1000 rpm for 10 s, then in the third step at 6000 rpm for 20 s. In the second step, a 500 µL of chlorobenzene solution was dripped just 8 s before the spin coating stops. The deposited samples were then annealed at 100 °C for 60 min. The detailed As prepared hole transport layer (HTL) of Spiro-OMeTAD solution making and spin-coating procedure was reported elsewhere.[33] Finally, around 100 nm Au metal electrode was thermally evaporated on top of the HTL layer to complete the device. The optimized thickness of anatase TiO$_2$ NPs (five cycle-coating) on FTO-substrate, perovskite layer, HTL, and Au layers were measured to be ~80, ~400, ~200, and ~100 nm, respectively.

4.5 Opto-electrical Simulation Method

A coupled optical-electrical 3D numerical simulation approach has been adopted to study the planar PSCs. Finite-difference time-domain (FDTD) optical simulations integrated with the particle swarm optimization (PSO) algorithm were carried to investigate the optics and optimization of the solar cells. The complex refractive indices of materials were used as input parameters for the FDTD simulation. The optical constants of materials were either determined through ellipsometry measurement or adopted from the earlier published work.[7] The complex refractive index of materials is provided in the Supplementary Information (Figure S16). The optical wave propagation for the device was modeled for a wavelength ranging from 300 to 800 nm close to the perovskite absorber bandgap. The incident plane wave is circularly polarized (TEM), which has an amplitude of 1000 mV/m. FDTD simulation
allows providing the electromagnetic field distribution, which further lets calculating power density, quantum efficiency (QE), and short-circuit current density (J_{SC}). A detailed discussion on how to calculate power density, QE, and J_{SC} is provided in the Supplementary Information (Section S1). Furthermore, the maximum value of the collection efficiency is expected to be unity since the perovskite thickness was considered in this study small than their diffusion length. Nevertheless, FDTD simulation does not provide the device's electrical parameter values, such as open-circuit voltage (V_{OC}), fill-factor (FF), which are essential to realizing realistic photovoltaic performance. Hence, the finite element method (FEM) simulations are carried out, which provide current-voltage (JV) characteristics of the device that further allow realizing V_{OC}, FF, and PCE of PSCs. The generation rate calculated from the optical simulation is used as a major input for the FEM simulation. More details on the FEM simulation can be found from the Supplementary Information (Section S2) and the literature.[7,62] Furthermore, a list of essential experimentally realized electronic properties of the material was used in the FEM calculation, selected wisely from the literature,[62–69] and values are itemized in the Supplementary Information (Table S4)

**Competing interests**

The authors declare no competing interests.

**Supporting Information**

Supplementary information is available from the authors.
Acknowledgments

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Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Mohammad Ismail Hossain, Ph.D. (equally contributed first author)

On behalf of all co authors
Highlights

Low-temperature Treated Anatase TiO$_2$ Nanophotonic-structured Contact Design for Efficient Triple-cation Perovskite Solar Cells

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Highlights of the Manuscript

- Low temperature processed high-quality TiO₂ nanoparticles film were prepared
- TiO₂ layer was optimized while fabricating efficient perovskite solar cells
- 3D opto-electrical simulations investigate optical and electrical properties of the device
- Efficient nanophotonic front contact is designed for efficient perovskite solar cells
• Optimized device enhances ECE by 25~30%, up to 23%, compared to the flat contact device