Enhancing solar cell efficiency: Investigation of high-performance lead-based perovskite-on-silicon (PVK–Si) tandem solar cells through design and numerical analysis

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Enhancing solar cell efficiency: Investigation of high-performance lead-based perovskite-on-silicon (PVK–Si) tandem solar cells through design and numerical analysis

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ABSTRACT
Perovskite solar cells have pulled off a level of conversion efficiency comparable to other well-established photovoltaics, such as silicon and cadmium telluride. Organic–inorganic halide perovskite materials are one of the most appealing and imminent options for developing high performance and cost-effective photovoltaic cells. In this simulation-based research, a highly efficient 2-terminal perovskite-on-silicon (PVK–Si) tandem configuration has been proposed with improved stability and significant cost savings. Initially, the MAPbI3 based perovskite top cell with 1.57 eV bandgap is tested with several distinct hole transport materials (HTMs) and electron transport materials (ETMs), four each. Spiro-OMeTAD as HTM and C60 as ETM are the best performing materials and employed in final top cell configuration to have a power conversion efficiency (PCE) of 23.05%. The systems for tandem configuration are experimented after setting the layers of individual top and bottom sub-cells to optimized thicknesses. The thicknesses of both top and bottom absorber layer are adjusted to find out the current matching point. The tandem configuration exhibits optimized thicknesses of 250 nm and 150 μm for top and bottom sub-cells, respectively. Performance evaluation of the top cell involves standard AM 1.5G solar spectrum illumination at a 250 nm absorber thickness, while the bottom cell’s performance is assessed using top cell filtered spectrum. The simulated tandem configuration composed of IZO/C60/MAPbI3/spiro-OMeTAD/n-nc-SiOx/n-c-Si/P+ Si showed a champion PCE of 35.31% with a fill factor of 79.46% and open-circuit voltage of 2.12 V. These findings signify substantial advancements in the field of PVK-Si tandem photovoltaic cells, marking a significant stride toward potential commercial applications.

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I. INTRODUCTION

Crystalline silicon (c-Si) is widely regarded as the most prominent material in photovoltaic (PV) cells, as it comprises nearly 90% of the photovoltaic market.1 Nevertheless, the benchmark conversion efficiency for silicon solar cells still remains at 26.7% after over three years,7 which is consistent with the theorized Auger limit of c-Si-based solar cells of 29.4%.8 Improving conversion efficiency appears to be a distant goal given the intrinsic losses associated with single-junction devices, such as thermalization and recombination losses. The multi-junction or tandem arrangement can be used to overcome this drawback by incorporating stacking materials with significantly reduced bandgaps from top to bottom. This allows for the absorption of photons with gradually declining energy values as
they travel through the cell structure. Two-terminal (2T) and four-terminal (4T) designs are the most prevalent types of designs.\textsuperscript{4} The mechanically stacked technique is necessary to produce the four exterior terminals for the second configuration, two external terminals from each sub-cell. Nonetheless, if the optical characteristics of individual sub-cells are optimized, the two-terminal arrangement may still offer certain efficiency improvements. c-Si-based tandem cells are reported to be capable of exceeding both the Auger limit and the Shockley–Queisser (SQ) limit for single-junction PV cells (33%).\textsuperscript{5} The first model of a halide perovskite solar cell was exhibited in 2009 and featured a conversion efficiency of 3.8%.\textsuperscript{6} A recent advancement in manufacturing processes facilitated the growth of perovskite materials with bandgaps ranging from 1.2 to 1.3 eV, resulting in a record 21% efficiency in the laboratory using a 4T tandem design.\textsuperscript{7}

In recent times, there has been a significant utilization of organic–inorganic metal halide perovskite materials in the development of advanced solar cells, aiming to enhance photovoltaic performance. These materials possess exceptional optical and electronic properties, including a strong spectral absorption, efficient light-harvesting competence, adjustable direct bandgap, long charge carrier span, diffusion lengths, and mobilities.\textsuperscript{8} The majority of research being conducted on perovskite solar cells is concentrated on materials with the structural formula $\text{ABX}_3$. The halide anion $\text{X}$, usually $\text{Cl}$, $\text{Br}$, or $\text{I}$, is combined with an inorganic cation $\text{B}$, such as $\text{Pb}$ or $\text{Sn}$. Additionally, the cation $\text{A}$ can be either an inorganic cation (usually $\text{Rb}$ or $\text{Cs}$) or an organic cation, such as $\text{MA}$ (methylammonium) or $\text{FA}$ (formamidinium).\textsuperscript{9} Despite the fact that multiple minerals hold the $\text{ABX}_3$ structure, only a handful are ideal for use as effective light concentrators in solar photovoltaics. The multi-junction or tandem arrangement and three-dimensional $\text{ABX}_3$ framework of this perovskite are shown in Figs. 1 and 2.\textsuperscript{10}

However, the most effective halide perovskite solar cells (PSCs) are made up of materials that contain lead. They have an acceptable and tunable bandgap range of 1.5–2.5 eV, excellent ability to absorb sunlight, optimal excitation energy, and moderately high mobility, all of which provide noble power conversion efficiencies, making them convenient for applications in the photovoltaic industry.\textsuperscript{11} Lead-based perovskites are exquisite semiconductors that have substantially all of the characteristics essential for long-term thin-film photovoltaics. The methylammonium lead halide ($\text{CH}_3\text{NH}_3\text{PbI}_3$), also recognized as $\text{MAPbI}_3$, is the most extensively utilized organic–inorganic halide perovskite.\textsuperscript{12} This form of perovskite gained popularity due to improving manufacturing methods and rapid advancements in conversion efficiency over a very short time span. They can be produced through low-cost, low-temperature solution processing techniques by utilizing elements that do not require energy-intensive cleaning.\textsuperscript{13} They can be readily produced through roll-to-roll printing methods at a much lower cost, which enhances their appeal for both large and small-scale grid integration. The crystal structure of $\text{ABX}_3$ provides significant flexibility within a wide range of possible compositions.\textsuperscript{14} They are made of a very thin active layer that requires only 1 g of perovskite material for each square meter of module.\textsuperscript{15} Even though they have excellent outcomes, the fact that the organic cations in perovskite are thermally unstable, hygroscopic, and volatile is a big worry for the large-scale marketing of perovskite photovoltaics.\textsuperscript{16} This difficulty has prompted efforts to enhance device stability, and a viable strategy in this regard is to incorporate novel materials, such as 2D perovskites, into 3D hybrid perovskites while still retaining excellent performance.\textsuperscript{17–19} In addition, several methods have been adopted to ensure better stability of 3D PSCs, with encouraging outcomes. These include device encapsulation, adapted crystallization, Pb-site doping, interfacial modulation, surface passivation, and structural engineering.\textsuperscript{20} In addition to developing the perovskite layer, the formation of effective charge-selective contacts is regarded as an important strategy for defining the system characteristics, given that the photo-induced carriers need to pass through the interface prior to reaching the electrodes.\textsuperscript{21} Numerous materials may be utilized in PSCs as electron transport layer (ETL) and hole transport layer (HTL) to enhance charge transport and inhibit charge recombination. Islam et al.\textsuperscript{22} calibrated a lead-based tandem solar cell with MAPbI$_3$-on-silicon utilizing the simulation software SCAPS 1D, yielding a power conversion efficiency (PCE) of 32.29% through extensive optimization of the absorber layer in terms of thickness and defect density.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{tandem_cell_structure.png}
\caption{Tandem cell structure with respective illuminations.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{structure_model.png}
\caption{Structure model of $\text{CH}_3\text{NH}_3\text{PbI}_3$.\textsuperscript{10} [Oku, Solar Cells: New Approaches and Reviews, 2015; licensed under a Creative Commons Attribution (CC BY 3.0) license.]}\end{figure}
However, the efficiency could be improved by optimizing the rest of the layers and reducing the defect density of the absorber layer slightly more. Shrivastav et al.1 utilized a tandem device of a perovskite-based top sub-cell and a c-Si-based bottom sub-cell with bandgap energies of 1.68 and 1.2 eV, respectively. Filtered spectra, current matching, thickness variation of the absorber layer, and illuminated J-V curves were carried out for tandem analysis that showcased a PCE of 33.05%. Madan et al.23 designed a 23.36% efficient 2T lead-sulfide-based perovskite-PbS CQD (colloidal quantum dot) tandem solar cell (TSC) through comprehensive simulations of the device to minimize the losses due to thermalization and transmission and to reduce cost. Although the long-term stability of lead-based PSCs is a massive limitation toward commercialization, the search for alternatives is still in the early stages of exploration.

In this work, a highly efficient perovskite-on-silicon tandem configuration is proposed with improved stability and significant cost-saving opportunities. Lead-based MAPbI3 with bandgap of 1.57 eV has been chosen as the halide perovskite material to be utilized in top cell absorber layer. Four different hole transport materials (HTMs) and four electron transport materials (ETMs) have been tested with the perovskite top cell to find out the materials with best performance. Each material utilized in the proposed PSC has been optimized in terms of thickness. The perovskite absorber layer is further optimized in terms of defect density. Each material has numerous advantageous features, such as excellent optoelectronic properties, improved chemical stability, and low-cost fabrication techniques. These efficient materials with appropriate thickness contribute to develop the optimal structure of proposed tandem configuration, resulting into a champion power conversion efficiency of 35.31%. This simulated performance exceeds both the Auger limit and the NREL record efficiency of 33.9% for perovskite/Si tandem cells.24 This work is also focused on improving device stability and lowering fabrication cost of the materials. It demonstrates details about the preparation of the materials and some novel approaches to significantly enhance their chemical stability. The implications of this research are the following:

- A highly efficient (35.31%) tandem solar cell of optimal structure.
- Use of chemically stable, cost-effective, and best performing materials.
- Valuable insights into the effect of different electron and hole transport materials on performance parameters.
- Thickness optimization, defect density optimization, filtered spectrum, and the current matching approach to simulate the tandem configuration.

II. DEVICE STRUCTURE AND SIMULATION METHODOLOGY

SCAPS-1D has been chosen throughout this work to do the solar cell simulation analyses due to its open-source nature, extensive capabilities, and user-friendly interface. SCAPS is a software to simulate one dimensional (1D) solar cell, which was originated at the University of Gent in Belgium by the Department of Electronics and Information Systems (ELIS).25 It supports solar cell simulation of up to seven layers of the semiconductor material. The single-cell standalone simulation can be done using SCAPS-1D inbuilt features. In this work, the standalone simulations have been done individually for both sub-cells by utilizing the standard AM1.5G 1 sun spectrum, and a widely adopted filtered spectrum has been utilized to simulate the tandem configuration following the current matching strategy.

A. Device structure of lead-based perovskite solar cells

The proposed perovskite-on-silicon (PVK-Si) tandem device structures comprised two sub-cells, namely, the top and bottom sub-cells, which combine a perovskite material of high bandgap on top of a c-Si material of low bandgap. Photons with high energy get captured by the layers of the top sub-cell, whereas photons with low energy travel through the top sub-cell to the bottom and get absorbed.27 Thus, tandem devices enable a larger photon absorbance than single junction devices, resulting in higher conversion efficiency. The layout of the top sub-cell consists of a front-contact, transparent conductive oxide (TCO) layer, electron transport layer (ETL), absorber layer (perovskite), hole transport layer (HTL), and back-contact, as shown in Fig. 3(a). Electron transport materials (ETM) extract and transport the negative charge carriers toward the electrical contact terminal as well as inhibit charge recombination by blocking the hole transport.28 On the contrary, hole transport materials (HTM) transport the holes toward the contact terminal and also inhibit charge recombination. In this study, the proposed top sub-cell layout has been experimented with four distinct electron transport materials, i.e., titanium dioxide (TiO2), phenyl-C61-butyric acid methyl ester (PCBM), compact tin oxide (SnO2)/PCBM, and buckminsterfullerene (C60) as the ETL. Likewise, for HTL, four different materials have been tested through device simulations, i.e., spiro-OMeTAD, PEDOT:PSS (poly(3, 4-ethylenedioxythiophene) polystyrene sulfonate), PTAA (poly(triaryl amine)), and Cu2O (copper oxide). The best performing materials have been selected for the final tandem configuration. The main absorber layer (perovskite) has been tested for a lead-based perovskite material, MAPbI3 (methyl ammonium lead iodide). The front contact (usually glass) and the back contact (usually metal) work as photo-electrodes and metal electrodes, respectively. The top sub-cell has been illuminated with the standard AM1.5G 1 sun spectrum, and thickness optimization has been performed to select the thicknesses for each layer used in the top sub-cell. In the case of the bottom sub-cell, the layout can be either an n-n-p or an n-p-p configuration. The emitter layer is composed of n-type semiconductor material, which inhibits the passage of holes while permitting the flow of electrons. The active layer can be constructed with either

![FIG. 3. Schematic diagram: (a) top cell configuration and the materials tested for ETL and HTL and (b) bottom cell configuration.](image-url)
In this study, the bottom sub-cell structures for standalone and tandem configurations comprised n-doped monocristalline silicon oxide (n-nc-SiOx), n-type crystalline silicon (n-c-Si), and highly doped p-type silicon layer (P+ Si), as shown in [Fig. 3(b)]. In the case of the PVK-Si tandem configuration [Fig. 4], the top sub-cell has been calibrated under the standard AM1.5G 1 sun spectrum, while the bottom device has been calibrated through simulations under the filtered spectrum of the top sub-cell.

### B. Characteristics of different materials used in the proposed design

#### 1. Electron transport material

This work has employed several electron transport materials, including TiO$_2$, PCBM, compact SnO$_2$/PCBM, and C$_{60}$ as ETLs in proposed PSCs considering their suitable energy levels and high electron mobility. TiO$_2$ is the one that is most often used of these ETLs. However, the fabrication of TiO$_2$ films necessitates a high-temperature operation above 450 $^\circ$C, which leads to higher cost of production and inhibits the progress of PSCs on flexible substrates. Hence, there is a high desire for the replacement of TiO$_2$ with ETLs produced at low temperature. C$_{60}$ has been employed as the ETL in this paper’s final tandem configuration as it outperformed the rest of the ETLs tested in this work, which is further discussed in the Result section. However, when C$_{60}$ is utilized as an independent ETL, the PSC efficacy degrades. Hence, it is often suggested to use a compound ETL consisting of solution-processed C$_{60}$ and any other compact electron transport materials, such as TiO$_2$, PCBM, and BCP (bathocuproine).

The C$_{60}$ film can be prepared on a TCO substrate by spin coating a solution of C$_{60}$ 1,2-dichlorobenzene at a speed of 2000 rpm for 30 s, followed by annealing at 60 $^\circ$C for 5 min on a hotplate. In recent times, solid-state ionic-liquids (IL) like 1-ethyl-3-methylimidazolium iodide ([EMIM]I) have been utilized to optimize solution-processed C$_{60}$ that exhibits improved thermal stability, good hydrophilic nature, and boosted ionic conductivity.

#### 2. Perovskite absorber (MAPbI$_3$)

We have selected widely employed methyl ammonium lead halide (MAPbI$_3$) with a bandgap of 1.57 eV as the primary absorber layer. This organic–inorganic lead halide perovskite has attained significant attention worldwide, thanks to its superior conversion efficiency, low cost, and easy fabrication methods. It exhibits an extended diffusion length, solid resistance to internal defects, and some distinctive optoelectronic features, including a linear tunable bandgap, ambipolarity, high charge carrier mobility, a sharp absorption edge, and simple synthesis. MAPbI$_3$ can be prepared in a cost-effective conventional laboratory through a wet chemical process and demonstrates significant durability when incorporated into 2D perovskite.

A two-step spin coating process can be employed to fabricate the perovskite (MAPbI$_3$) thin film, in which a solution of methyl ammonium iodide (MAI) is usually spin-coated on a film of lead iodide (PbI$_2$). In the first stage, a solution of PbI$_2$ (0.4 g in 2 ml of N,N-dimethylformamide, or DMF) is applied onto the HTL surface using spin coating at a speed of 1000 rpm for 10 s, followed by annealing at 100 $^\circ$C for 10 min. In the later stage, 0.01 g of MAI solution mixed with 2 ml of DMF is spin coated onto the PbI$_2$ film at 500 rpm for 5 s and 1000 rpm for 10 s. The film is then heated to 100 $^\circ$C for 10 min and the light-yellow color of perovskite film will be transformed into dark brown.

#### 3. Hole transport material

Highly efficient hole transport layers (HTLs) serve a dual purpose in facilitating hole transportation and impeding electron flow, while also functioning as a protective barrier against the detrimental effects of oxygen and moisture on perovskite films. Several hole transport materials, i.e., spiro-OMeTAD, PEDOT:PSS, PTAA, and Cu$_2$O, have been tested in to be utilized as HTL in proposed PSCs of this study. Among these, the spiro-OMeTAD compound has emerged as the leading hole-transporting material (HTM) in the field of highly efficient PSCs. This is primarily attributed to its favorable energy level, mild glass-transition temperature, simplicity of use, precise performance, and exceptional ability to be solution-processed.

To prepare the precursor solution for spiro-OMeTAD fabrication, an initial mixer is prepared by combining 17.5 ml of bis(trifluoromethylsulfonyl)imide lithium salt (LiTFSI solution, 0.55 Mm in acetonitrile), 28.6 ml of 4-tert-butylpyridine (tBP with purity of 98%), and 75.0 mg of spiro-OMeTAD. This mixture is then stirred for 3 h in 1 ml of chlorobenzene (CB) with a purity of...
99.8%. This precursor solution is then spin-coated onto the perovskite layer to fabricate the spiro-OMeTAD film at a speed of 5000 rpm for 30 s. A novel Spiro-OMeTAD synthesis employs green chemistry techniques, such as solventless, micellar, and water-based processes to reduce hazardous chemicals and waste while preserving the integrity and efficacy of the material. This approach effectively mitigates environmental impact and cost of synthesis. The purification method employs minimal organic solvents, yielding higher purity material and enhancing cell performance in terms of durability and reproducibility compared to existing commercial samples.

C. Simulation strategy

The current matching approach has been employed to combine both sub-cells in tandem configuration. The filtered spectrum at any perovskite thickness can be calculated following Eq. (1),

\[ S(\lambda) = S_0(\lambda) \times \exp\left(\sum_{i=1}^{n} a_{mat,i}(\lambda)d_{mat,i}\right), \]

where \( S(\lambda) \) represents the intensity of the filtered spectrum by the top sub-cell, \( S_0(\lambda) \) is the incident light intensity (W/m²) for the conventional AM1.5G 1 sun spectrum, \( a_{mat,i} \) symbolizes the absorption coefficient of the respective material, \( d_{mat,i} \) represents the thickness, and the subscript, mat, represents any particular material (\( i = 1 \) for TCO materials, 2 for ETL materials, 3 for perovskite absorber materials, and 4 for HTL materials). The absorption coefficients for each material are accessed from the SCAPS layer properties panel. In tandem configurations, current must flow equally through the sub-cells (top and bottom) that are connected in series. The tandem current density \( (I_{SC}) \) is limited by the standalone cell that has the lower current density, whereas net tandem \( V_{OC} \) is determined by adding the individual \( V_{OC} \) of the devices connected. The filtered spectrum, as shown in Fig. 5, has been created by varying the perovskite layer thickness to find out the current matching point for both sub-cells to connect in the tandem arrangement. The fill factor (FF) can be calculated as the ratio of maximum power \( (P_{max} = I_{mpp}V_{mpp}) \) to the product of \( V_{OC} \) and \( J_{SC} \) of the solar cell experimented following Eq. (2). The subscript “mpp” designates the maximum power point along the J-V characteristic at which the solar cell delivers the maximum amount of power,

\[ FF = \frac{I_{mpp}V_{mpp}}{J_{SC}V_{OC}}. \]

The power conversion efficiency is derived by calculating the ratio of maximum generated power to the incident power on the solar cell tested [Eq. (3)]. The incident light is distinguished by the standard AM1.5G spectrum featuring an intensity of \( I_{in} = 1000 \) W/m²,

\[ \eta = \frac{P_{max}}{I_{in}} = \frac{I_{mpp}V_{mpp}}{I_{in}} = \frac{I_{oc}V_{oc}FF}{I_{in}}. \]

The single-junction standalone simulation can be done using the SCAPS-1D inbuilt features. Figure 6 depicts a flow chart of the standalone simulation procedure in SCAPSD software. Initially, the working conditions (temperature, voltage, and frequency), illumination conditions, and resistances are defined for the proposed solar cell to be simulated. Finally, the simulation is run by calculating single shot on SCAPS interface, which provides the results of calculation.

D. Simulation parameters

A comprehensive literature survey has been carried out in order to assess the electrical characteristics of all of the materials utilized in the solar cell structures for numerical analysis. Table I contains the chosen parameters for HTL and ETL, while Table II contains the parameters for the TCO layer, MAPbI₃, and the layers of the bottom sub-cells. The defect type of each layer has been selected as neutral and the energetic distribution for each defect have been taken as single where both the cross-sectional area of electrons and holes are taken as \( 1 \times 10^{-19} \) cm². The energy level relative to the reference defect energy level is 0.6 eV. The uniform defect densities for each layer have been defined using the defect properties panel of SCAPS 1D.

III. RESULT AND DISCUSSION

Both the top and bottom sub-cells have undergone standalone simulation, and the current matching strategy has been adopted for simulating the tandem configuration. The top sub-cell layout was tested for different HTMs and ETMs through a series of simulation.
The numerical performance was evaluated against various thicknesses for each layer, and optimized thicknesses were chosen for further simulation study in this research. Consequently, the defect density of the perovskite layer (MAPbI$_3$) had also been optimized. In the case of bottom sub-cell, an optimized structure was chosen for both the standalone and tandem simulations. Simulation results include the performance parameters of a solar cell (single-junction or tandem), i.e., open-circuit voltage ($V_{OC}$), short circuit current density ($J_{SC}$), fill factor (FF), and power conversion efficiency (PCE).

### A. ETL variation of top sub-cell

The materials tested for ETL are TiO$_2$, PCBM, SnO$_2$/PCBM compact ETL, and C$_{60}$. While testing materials for ETL, the hole transport layer had been chosen as Spiro-OMeTAD (1 nm) and back contact. The performance of each ETMs (V$_{OC}$, J$_{sc}$, FF, and PCE) has been calibrated using SCAPS batch and recorder panel for thickness of 10–460 nm in ten steps, as shown in

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Top Sub-cell</th>
<th>Bottom Sub-cell</th>
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</thead>
<tbody>
<tr>
<td>Bandgap (eV)</td>
<td>TiO$_2$</td>
<td>n-nc-SiOx</td>
</tr>
<tr>
<td>3.5</td>
<td>1.4</td>
<td>1.12</td>
</tr>
<tr>
<td>Electron affinity (eV)</td>
<td>PEDOT:</td>
<td>n-c-Si</td>
</tr>
<tr>
<td>4.5</td>
<td>3.9</td>
<td>4.05</td>
</tr>
<tr>
<td>Dielectric (relative) permittivity</td>
<td>TiO$_2$</td>
<td>P + Si</td>
</tr>
<tr>
<td>10</td>
<td>11.9</td>
<td>11.9</td>
</tr>
<tr>
<td>CB, effective density of states (cm$^{-3}$)</td>
<td>SnO$_2$/PCBM</td>
<td>1400</td>
</tr>
<tr>
<td>1.9 x 10$^{19}$</td>
<td>1.4 x 10$^3$</td>
<td>1.4 x 10$^3$</td>
</tr>
<tr>
<td>VB, effective density of states (cm$^{-3}$)</td>
<td>SnO$_2$/PCBM</td>
<td>1400</td>
</tr>
<tr>
<td>1.9 x 10$^{19}$</td>
<td>2.8 x 10$^{19}$</td>
<td>2.8 x 10$^{19}$</td>
</tr>
<tr>
<td>Thermal velocity, electron (cm/s)</td>
<td>SnO$_2$/PCBM</td>
<td>1400</td>
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<tr>
<td>10$^7$</td>
<td>7.6 x 10$^7$</td>
<td>7.6 x 10$^7$</td>
</tr>
<tr>
<td>Thermal velocity, hole (cm/s)</td>
<td>SnO$_2$/PCBM</td>
<td>1400</td>
</tr>
<tr>
<td>7 x 10$^7$</td>
<td>1.4 x 10$^5$</td>
<td>1.4 x 10$^5$</td>
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<tr>
<td>Shallow uniform donor density, N$_D$ (cm$^{-3}$)</td>
<td>SnO$_2$/PCBM</td>
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<tr>
<td>1 x 10$^{18}$</td>
<td>1 x 10$^{20}$</td>
<td>1 x 10$^{20}$</td>
</tr>
<tr>
<td>Shallow uniform acceptor density, N$_A$ (cm$^{-3}$)</td>
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<tr>
<td>0</td>
<td>0</td>
<td>0</td>
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<tr>
<td>Defect density, N$_i$ (cm$^{-3}$)</td>
<td>SnO$_2$/PCBM</td>
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<td>References</td>
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Figs. 7(a)–7(d). It can be seen from the figures that C₆₀ as ETL has outperformed the other materials in terms of PCE, Jₛ𝑐, and FF values. In the case of Vₒ𝑐, the compact ETL of SnO₂/PCBM has shown the best of values although the values of Vₒ𝑐 can be seen changed very little with respect to the change in thickness.

So, C₆₀ was chosen as the suitable electron transport material to be used as ETL in both standalone and tandem structures. It is evident that the conversion efficiency increases up to a thickness of 200 nm of C₆₀ and then it remains almost constant after any further increase. The thickness for C₆₀ was selected as 150 nm in order to keep it thin so that it does not get larger than the absorber layer itself. TiO₂ is also a widely used electron transport material due to its higher conversion efficiency, but the stability issues of it still remains a challenge. The use of C₆₀ as ETL has recently gained popularity due to its performance and much improved UV stability. It is often suggested to use a compact ETL of C₆₀ and any other electron transport materials, such as TiO₂, PCBM, and BCP (bathocuproine), to facilitate the fabrication of C₆₀ films and to enhance the blocking of charge recombination holes at the interface between ETL and perovskite.

B. HTL variation of top sub-cell

In a similar manner, the structure of top sub-cell has been tested for four different HTMs, i.e., spiro-OMeTAD, PEDOT:PSS, PTAA, and Cu₂O, to be utilized as HTL. While testing materials for HTL, the electron transport layer had been chosen as C₆₀ (150 nm) as it was optimized before and the rest of the layers had been kept unchanged. So, the structure considered for this time comprises front contact/IZO (40 nm)/C₆₀ (150 nm)/MAPbI₃ perovskite (600 nm)/HTL and back contact. The performance parameters of each HTMs have been calibrated using SCAPS batch and recorder panel for thickness of 1–100 nm in ten steps, as shown in Figs. 8(a)–8(d). It is apparent from Fig. 8 that the values of PCE and fill factor are almost similar for each HTLs tested except Cu₂O. In the case of Vₒ𝑐 and Jₛ𝑐, the impact can be considered negligible as they vary within a very close range under varying thicknesses. After thorough analysis, spiro-OMeTAD of 1 nm has been chosen as the suitable hole transport material to be used as HTL in both standalone and tandem configurations as it is widely utilized in solar industry and performs well at lower thickness.

C. Optimization of thickness and defect density

The standalone simulation of top sub-cell has been optimized in terms of thickness of each layer. In Secs. III A and III B, the ETL and HTL have already been optimized, and in this segment, the impact of thickness on PV characteristics has been observed for rest of the layers, i.e., IZO and MAPbI₃ perovskite absorber. An optimized thickness has been chosen for each layer based on the best performance through comprehensive analysis. The perovskite absorber layer has also been optimized in terms of defect density. Figure 9(a) shows the PV parameters for thickness variation of IZO layer (10–460 nm in ten steps).

**FIG. 7.** The performance of various ETMs tested in standalone top sub-cell simulation.
It can be seen that variation in thickness has a very little impact on the outcomes. The optimal thickness of IZO has been chosen to be 40 nm as a decrease in thickness results in better conversion efficiency and fill factor. Similarly, the absorber perovskite layer (MAPbI$_3$) has been calibrated for thickness variation from 100 to 1000 nm [Fig. 9(b)]. It is apparent that with increasing thickness, the open-circuit voltage and fill factor decrease, but the conversion efficiency and current density increase, as expected. A perovskite layer thickness of 600 nm has been selected as optimal to obtain the best performance of top sub-cell in standalone configuration.

In addition, the top sub-cell has also been optimized with regard to defect density of perovskite layer (from $10^{13}$ to $10^{14}$ cm$^{-3}$ in ten steps), as seen in Fig. 9(c).

It is obvious that a reduction in defect density leads to an increase in higher power conversion efficiency and fill factor. However, the defect density should not be considered much lower just to obtain higher PCE as it is impossible to find an ideal solar cell with no defects. Initially, the defect density of MAPbI$_3$ was considered as $7 \times 10^{15}$ cm$^{-3}$ following the work$^{41}$ and later, after analyzing the results of defect density variation, the optimized value for perovskite layer has been chosen as $1 \times 10^{13}$ cm$^{-3}$.

### D. Standalone simulation of top and bottom sub-cells

The final layout of top sub-cell with selected layers after thickness and defect density optimization comprised front contact/IZO (40 nm)/C$_{60}$ (150 nm)/MAPbI$_3$ (600 nm)/spiro-OMeTAD (1 nm) and back contact. The bottom cell structure considered for standalone configuration is comprised of n-nc-SiO$_x$ (0.095 μm)/n-c-Si (150 μm)/ and P$^+$ Si (10 μm). The standalone simulation for both sub-cells have been performed under standard AM1.5G 1 sun spectrum and the characteristics J-V curve with performance parameters for each configuration are shown in Figs. 10(a) and 10(b).

### E. Tandem cell simulation

To simulate the lead-based PVK-Si tandem structure, the top sub-cell was simulated using the prevalent AM1.5G-1 sun spectrum, while the simulation of the bottom sub-cell was performed under the filtered spectrum of the top cell following the current matching strategy. To connect the sub-cells in tandem, current must flow equally through the cells connected. Therefore, the thickness of the absorber layers of both devices has been adjusted simultaneously to determine the optimal thickness that satisfies the current matched situation aforesaid. The top sub-cell has been simulated...
FIG. 9. Impact on PV parameters: (a) thickness optimization for the IZO layer, (b) thickness optimization of the MAPbI3 layer, and (c) defect density optimization of the MAPbI3 layer.
FIG. 10. J-V characteristics curve with other performance parameters: (a) top sub-cell and (b) bottom sub-cell.

under a standard spectrum with thickness variation of the perovskite layer from 60 to 600 nm (in ten distinct increments), and the bottom cell has been simulated under filtered spectra of the respective perovskite layer thicknesses. Thereafter, the current matching point at which the total amount of current flowing through the top and bottom devices is the same has been determined by plotting a current matching curve, as illustrated in Fig. 11. The current matching curve shows that the top sub-cell with a perovskite layer of 250 nm and the bottom sub-cell with an active layer of 150 μm give the same current density value (20.96 mA/cm²) for standalone simulations. Hence, a tandem configuration can be simulated following this current matching point.

The J-V characteristics curves of top and bottom cells were individually calculated and summed, where voltages at constant currents were added in order to generate the tandem J-V characteristics curve of lead-based PVK-Si tandem cell. It has been determined that the open-circuit voltage of the tandem configuration is 2.12 V and the short-circuit current density (Tandem J_{SC}) is 20.96 mA/cm². Later, the power curve (P–V) has also been plotted to find the

FIG. 11. Current matching curve for the top and bottom sub-cells with variable absorber layer thicknesses.

FIG. 12. J-V characteristics curve and power curve for the lead-based tandem solar cell.

FIG. 13. J-V characteristics curves of each simulated device, both standalone and tandem configuration.
maximum power point (mpp) and the results have been found as $V_{\text{mpp}} = 1.78 \text{ V}$ and $J_{\text{mpp}} = 19.83 \text{ mA/cm}^2$ and resulting into maximum power of 35.31 mW/cm$^2$. The characteristics curves of PVK-Si tandem cell are illustrated in Fig. 12.

The J-V curves for each individual cell (both standalone and tandem configuration) are displayed in Fig. 13. The simulated performance parameters for the devices calibrated in this work are shown in Table III, and the study has been compared with previously published performance results of lead-based PVK-Si tandem solar cells in renowned literature, as shown in Table IV. It can be seen that the conversion efficiency for the lead-based PVK-Si tandem solar cell in this study has outperformed the rest of the devices mentioned above. This is because the best-performing materials have been utilized in the structures of this study through optimization of thickness and defect density, which has yielded greater conversion efficiency. Moreover, the series and shunt resistance considered in this work also have a great influence on the performance, which was not clarified in the other literature mentioned herein. Comparing the results of this study with other published literature, the simulation methodology and tandem devices considered in this work can be justified as a promising option for future solar cell technology with further advancements and emphasis.

### IV. CONCLUSION

To conclude, we have simulated a perovskite/Si tandem solar cell with an outstanding power conversion efficacy of 35.31%. The two-terminal (2T) tandem configuration is composed of a MAPbI$_3$-based top cell of high bandgap and a crystalline-silicon based bottom sub-cell of low bandgap. The standalone conditions of both sub-cells are simulated under the illumination of AM1.5G solar spectrum to have power conversion efficiency of 23.05% and 23.65%, respectively. In tandem configuration, the top cell is illuminated with standard solar spectrum, while the bottom cell is simulated under the filtered spectrum following the current matching strategy. The current matching is attained at a thickness of 250 nm for perovskite absorber and 150 µm for bottom cell, resulting in a tandem short circuit current density of 20.96 mA/cm$^2$. The net tandem open-circuit voltage ($V_{\text{OC}}$) is calculated by adding the voltages across the individual cells at current matching point. The tandem configuration composed of IZO/C$_{60}$/MAPbI$_3$/spiro-OMeTAD/n-nc-SiOx/n-c-Si/P+ Si showed a fill factor of 79.46% and $V_{\text{OC}}$ of 2.12 V. In the future, our focus lies in enhancing the long-term stability of PSCs against heat and moisture, a crucial step to facilitate commercialization of PSCs. This can be achieved by integrating new materials, such as 2D perovskites or mixed cations/halides, and doping certain metals, such as Cs, into perovskite materials.

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### AUTHOR DECLARATIONS

#### Conflict of Interest

The authors have no conflicts to disclose.

#### Author Contributions

Nibir Mondol: Conceptualization (lead); Formal analysis (lead); Methodology (lead); Project administration (lead); Supervision (lead); Validation (lead); Writing – original draft (lead); Writing – review & editing (lead). Kazi Siamul Islam: Conceptualization (lead); Data curation (lead); Formal analysis (lead); Methodology (lead); Resources (lead); Software (lead); Validation (lead); Writing – original draft (lead). Md Rafiquel Islam: Conceptualization (lead); Investigation (lead); Methodology (lead); Project administration (lead); Resources (lead); Supervision (lead); Visualization (lead); Writing – review & editing (lead). Sajal K. Das: Conceptualization (lead); Data curation (lead); Formal analysis (lead); Investigation (lead); Project administration (lead); Supervision (lead); Visualization (lead); Writing – review & editing (lead).
DATA AVAILABILITY
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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