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Chapter

Advanced Developments in Fabrication Techniques and Bandgap Engineering in Perovskite Solar Cells (PSC)

*Lyle A. September, Ntombizonke Kheswa, Ntalane S. Seroka
and Lindiwe Khotseng*

Abstract

Perovskite materials have attracted attention because of their favorable features, which include configurable bandgap, improved charge transport, and unique electronic structure. The authors emphasize the most recent developments in PSC synthesis, innovative device architecture, bandgap engineering, charge transport processes, and stability. The need for a comprehensive design principle that surpasses the iterative process of the current synthesis of perovskite materials is highlighted. This is essential for developing high-quality, defect-free crystal structures with controlled dimensionality, and the choice of the deposition process is crucial to improve structural morphology in the production of PSC. This chapter also highlights that tandem and hybrid perovskite research is still in early development and emphasizes the need for basic research to fully understand the optical, electrical, and physical characteristics of the material before advances in industry.

Keywords: perovskite solar cells, two-step perovskite deposition, bandgap engineering, optoelectronic properties, material stability

1. Introduction

Finding safe, dependable, and clean energy sources is aided by the steady rise in technological innovation, global warming, and rising living standards. Because of this, conventional fossil fuel energy sources are insufficient to maintain both environmental preservation and the sustainable expansion of human society [1]. Consequently, there is a great deal of momentum behind rigorous scientific research to find sustainable renewable energy, with one of the most practical, dependable, and inexhaustible resources being solar energy. In general, a solar cell is an apparatus that converts light energy directly into electrical energy through photovoltaic effects or photochemical reactions. Perovskite solar cells have garnered a lot of attention lately due to their exceptional photovoltaic capabilities [2].

Optoelectronic device research has been thoroughly investigated in developing solar photovoltaic materials throughout the last few decades. The current rush in research and commercial attention is due to the critical role that perovskites play in the solar energy industry, with progressive properties in high power conversion efficiency (PCE) and low fabrication cost [3]. The rapid advance in efficiencies of perovskite/silicon (Si) tandem solar cells and metal halide semiconductor materials shows significant improvements in perovskite technology. Recent advances in metal halide semiconductors have been acknowledged as one of the most interesting possible light-absorbing materials with remarkable charge carrier transport performance in optoelectronic materials, particularly for photovoltaic applications. Optoelectronic devices based on metal halide semiconductors have increased developments in perovskite device performance [4, 5].

Perovskite solar cells (PSCs) have garnered a lot of attention from researchers lately because of their outstanding optical and electrical features, ease of fabrication, low production costs, and increased efficiency. As shown in **Figure 1**, recent studies have demonstrated an increase in efficiency and that monolithic perovskite/Si tandem cells can exceed the Shockley-Queisser limit for silicon, yielding a record of 33.7% created by KAUST. As single-junction silicon photovoltaics (PVs), which currently dominate the industry, get closer to their theoretical efficiency limit of 29%. Hybrid organic-inorganic perovskites (HOIPs) are one of the most well-liked next-generation solar materials due to their remarkable certified power conversion efficiency (PCE), which exceeds 25%. The National Renewable Energy Laboratory (NREL) has confirmed that Longi Green Energy, a Chinese photovoltaic manufacturer, achieves maximum efficiency. Considering the remarkable power conversion efficiency that has been achieved thus far, the main barriers to PSC commercialization are material selection, the fabrication processes used, band gap engineering, degradation, hysteresis effect, and stability problems [6]. These notable challenges are motivating academics to create efficient PSCs.

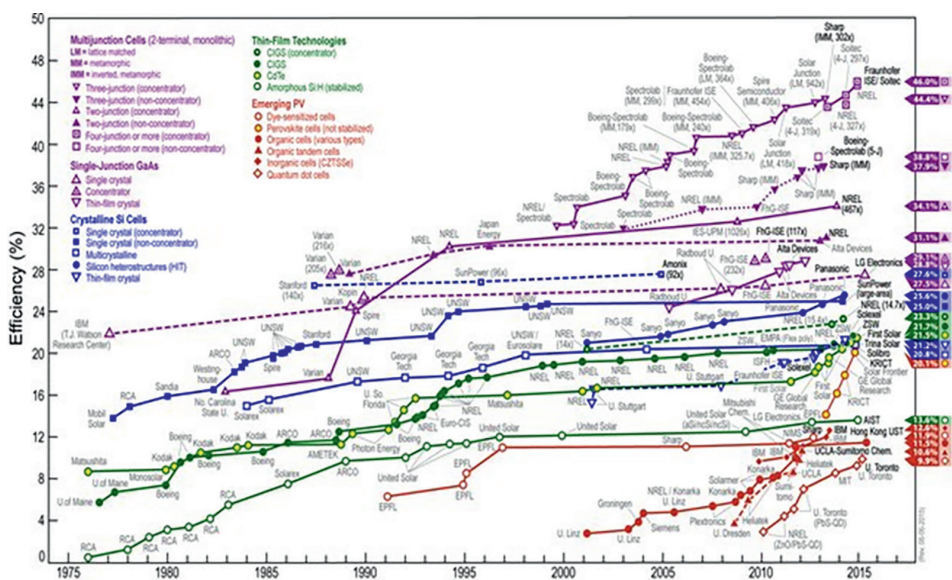


Figure 1. Best solar cell efficiencies reported by the National Renewable Energy Laboratory (NREL).

2. Recent advances

2.1 Background

PSCs generally have the formula ABX_3 , where X is a halogen anion, B is a divalent cation (Pb^{2+} or Sn^{2+}), and A is a monovalent organic or inorganic cation, most often methylammonium (MA^+), potassium (K), or cesium (Cs), illustrated in **Figure 2** [7]. The architecture of PSCs is also essential to their effectiveness because carrier generation and extraction depend heavily on the morphology of the various layers and their interfacial characteristics. PSCs are typically created by stacking perovskite active layers, anode/cathode, and carrier transport layers (also known as electron/hole transport layers, or ETLs/HTLs, respectively) to create a multi-interface system [8]. Based on a unique crystal structure called perovskite, perovskite solar cells absorb light by employing substances that are hybrid organic–inorganic, such as tin halide or lead halide. Electrons in the perovskite layer are excited by sunlight, which results in the creation of electron–hole pairs, or excitons. At the interfaces where the perovskite layer joins adjacent charge transport layers—such as titanium dioxide for electrons and spiro-OMeTAD for holes—efficient charge separation takes place [9]. Electrodes at the cell's contacts then gather electrons and holes, which when they travel *via* an external circuit, produce an electric current. Electrical power can be produced from this current.

The unique characteristics of perovskite materials, such as their high carrier diffusion length (greater than $1\ \mu m$), high electron mobility ($800\ cm^2/Vs$), and electrical characteristics (charge transport behavior), make them one of the best materials for the next generation of photovoltaic technology [10]. A multitude of different components can be combined to generate perovskite structures. Its features can be altered by modifying its composition and structure. To assess the stability of the perovskite structure, the parameters “tolerance factor” (t) and “octahedral factor” (u) should be considered during the design process. The perovskite's t and u range from 0.78 to 1.05 and 0.44 to 0.9, respectively, when it is in a stable structure [11].

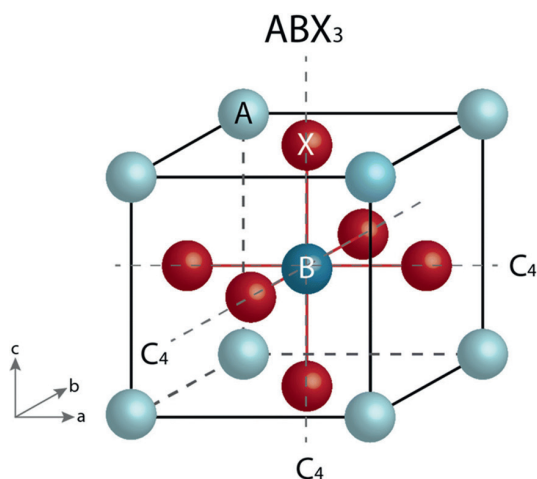


Figure 2.

The perovskite crystal structure with the molecular formula of ABX_3 (A: Cs^+ , Rb^+ , $MA^+ = (CH_3NH_3)^+$, or $FA^+ = (CH(NH_2)_2)^+$; B: Pb^{2+} or Sn^{2+} ; and X: Cl^- , Br^- , or I^-) Liao et al. [7].

The lack of stability studies in current perovskite-based cell research trends limits its applicability in outdoor applications. There would be very few commercial uses for perovskite solar cells if research on their stability and rate of degradation was not conducted. According to recent research on PSCs, chemical stability is greatly influenced by oxygen, temperature, moisture, UV light, and solution processing techniques [12]. PbI_2 and methylammonium iodide (MAI), for example, react very easily at low temperatures when driven by hydrogen and ionic interactions, making the synthesis of perovskite surprisingly straightforward experimentally [13].

Perovskite solar cells (PSCs) have advanced rapidly, with remarkable performance uptake from 3.8% to >26%; yet, the main obstacles to their commercialization are their low stability and toxicity. Many fabrication techniques and changes have been investigated to date in order to enable its commercialization [14]. The excellent light absorption across a wide spectrum, a possibility for low-cost manufacture by solution processing, and the theoretically high efficiency of perovskite solar cells make them an attractive technology [15]. The goal of ongoing research is to make perovskite solar cells a viable commercial alternative to silicon solar cells by improving stability and addressing environmental issues like lead toxicity in specific compositions [16].

2.2 Materials

Perovskite materials have attracted a lot of attention due to their unique optical, thermal, and electromagnetic capabilities, as well as their cubic lattice-nested octahedral structures. Perovskite materials have the following four characteristics: these materials have strong optical absorption coefficients, good photoelectric properties, and low exciton binding energies [17]. Perovskites have a high dielectric constant that facilitates efficient electron and hole photogeneration; furthermore, they can absorb solar energy effectively while successfully transporting electrons and holes from 100 nm to more than 1 μm . When employed in solar photovoltaic applications, these perovskite materials provide noticeably greater open-circuit voltages and short-circuit current densities [18]. Perovskite materials can also be classified as lead-free perovskites, all-inorganic perovskites, and organic–inorganic hybrid perovskites based on their chemical composition.

To further understand PSC performance and potential, a few perovskite materials are described. **Figure 3** describes the charge transfer and energy levels in flexible perovskite solar cells (F-PSC), because of a lower exciton binding energy, the photo-generated electron–hole pairs in the perovskite layer dissociate readily to free carriers [20]. Furthermore, the transparent conductive plastic substrates are coated with the ETL, which functions as a charge-selective layer. A perovskite absorption layer is then created using a spin-coating process. After that, the back electrodes and the HTL are deposited to create a normal n-i-p structure planar F-PSC. Additionally, the charge transport layers' locations are reversed for an inverted p-i-n planar F-PSC [19].

Many materials have been developed for use as ETL, HTL, and active layers in PSCs, including SiO_2 , TiO_2 , CdS , ZnO , PEDOT:PSS, NiO_x , Spiro-OMeTAD, PCBM, MAPbI_3 , MAPbBr_3 , MASnI_3 , CsPbBr_3 , FASnI_3 , and so on [21]. Specifically, Spiro-OMeTAD, the hole-transporting material used in perovskite solar cells, is more expensive and needs a labour-intensive production procedure [22]. New low-cost hole transport materials must be developed in order to support the commercialization of perovskite solar cells. The methylammonium lead iodide (MAPbI_3 — $\text{CH}_3\text{NH}_3\text{PbI}_3$) perovskite solar cell's favorable optical and electrical absorption coefficients contribute to its exceptional performance. Furthermore, the MAPbI_3 absorber layer has one

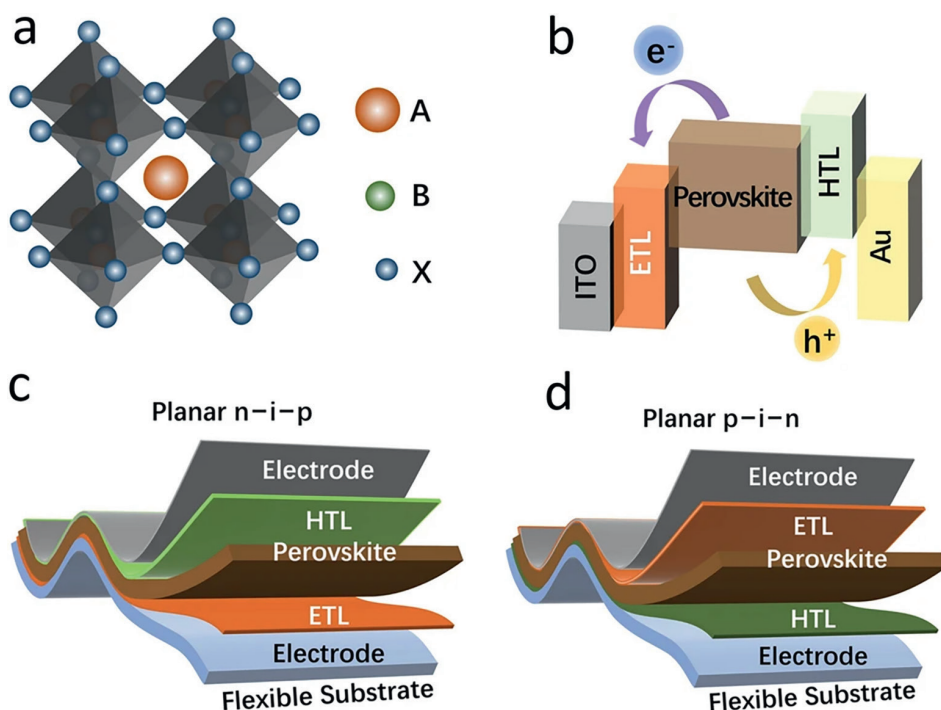


Figure 3. (a) Diagram showing the typical crystalline structure of ABX_3 perovskite. (b) Diagram showing the relative energy levels and charge transmission. Typical flexible PSC devices with $p-i-n$ and $n-i-p$ architectures, (c) and (d), respectively [19].

of the highest open-circuit voltages (V_{oc}), the lowest V_{oc} loss/deficit, and the lowest exciton binding energy [23]. These characteristics lead to improved charge transport, respectable charge carrier mobilities, and extended charge carrier diffusion lengths, which make it a viable option for photovoltaic applications. The $MAPbI_3$ material-based PSC has a very good PCE of about 23.7%, an acceptable band gap of 1.55 eV, remarkable electron mobility of $24.0 \pm 7 \text{ cm}^2 \text{ v}^{-1} \text{ s}^{-1}$, and hole mobility of $105 \pm 35 \text{ cm}^2 \text{ v}^{-1} \text{ s}^{-1}$ [24]. The most extensively studied perovskite solar cell is $MAPbI_3$ -based, and it has a wide range of possible applications.

Similarly, other materials such as $FAPbI_3$ and $CsPbI_3$ are utilized. The 1.43 eV narrow bandgap of $FAPbI_3$ is within the optimal range for achieving very high single-junction solar cell efficiency. Furthermore, compared to $MAPbI_3$, $FAPbI_3$ has superior semiconducting characteristics, such as a lower electron-photon coupling, a longer carrier lifetime, and a smaller charge carrier effective mass. Furthermore, the MA escape-induced device deterioration of $MAPbI_3$ results in low thermal stability. Additionally, formamidinium lead iodide ($FAPbI_3$)-based perovskite solar cells have a 19.09% efficiency at room temperature. The $FAPbI_3$ material has a narrow band gap, a high light-absorption coefficient, and excellent photo- and thermal-stability [25]. However, PSCs will find the small band gap of $FAPbI_3$ perovskites to be very beneficial, which may speed up the commercialization of this fascinating perovskite material.

Furthermore, the industrial advancement and growth of PSCs will be hindered by the exceedingly hazardous nature of the Pb element utilized in perovskite solar

cells. Since Pb is one of the toxic lead components in the MAPbI₃, replacing Pb with safer elements such as tin and germanium is the usual technique for eliminating toxic lead elements. Consequently, a low-toxicity or nontoxic component needs to be discovered in the future to replace Pb [26]. However, compared to PSCs using lead-based perovskites, PSCs using these alternative metal-based perovskites perform worse. This is due to the fact that the deterioration of perovskite devices has been linked to the migration of mobile ions [27]. As an example, there are two main problems that hinder the performance of PSC based on CsPbI₃. The low tolerance factor causes the preferred phase transition from the perovskite to the non-perovskite phase. Energy loss from energy-level imbalances and inevitable failures is the second, and it limits PCE [28].

Despite the superior performance of lead-based PSCs, lead toxicity necessitates the development of lead-free PSCs, shown in **Figure 4**. However, Pb exposure can be reduced by developing effective encapsulating methods. At the end of the module's lifecycle, it is imperative to ensure that all materials have been effectively recycled [29]. The large-scale production and commercialization of lead-based PSCs could release lead emissions into the environment, which could have a detrimental effect on the environment. Using a two-step spin coating technique, a low-toxicity perovskite material with 50% Pb and 50% Sn was produced while considering the toxicity and environmental effects of Pb [30]. The PSCs display an efficiency of 13.6% while utilizing Rs 5.3 Ω cm² and the device configuration ITO/PEDOT: PSS/CH₃NH₃Sn_{0.5}Pb_{0.5}I₃/C₆₀/Ag [22].

With a PCE of 29.74% and an ideal band gap of 1.3 eV, the FTO/TiO₂/CH₃NH₃SnI₃/CuSbS configuration, a tin (Sn) halide perovskite device, exhibits improved absorption in the visible spectrum area [32]. Nevertheless, unlike lead perovskite, tin halide perovskite readily oxidizes to the oxidation state Sn⁴⁺, resulting in metallic properties and somewhat limited air stability. Similarly, germanium iodide perovskite (MAGeI₃), a distinct group 14 metal with significant promise for use in solar applications, is the most obvious substitute for lead in monovalent applications. The low performance

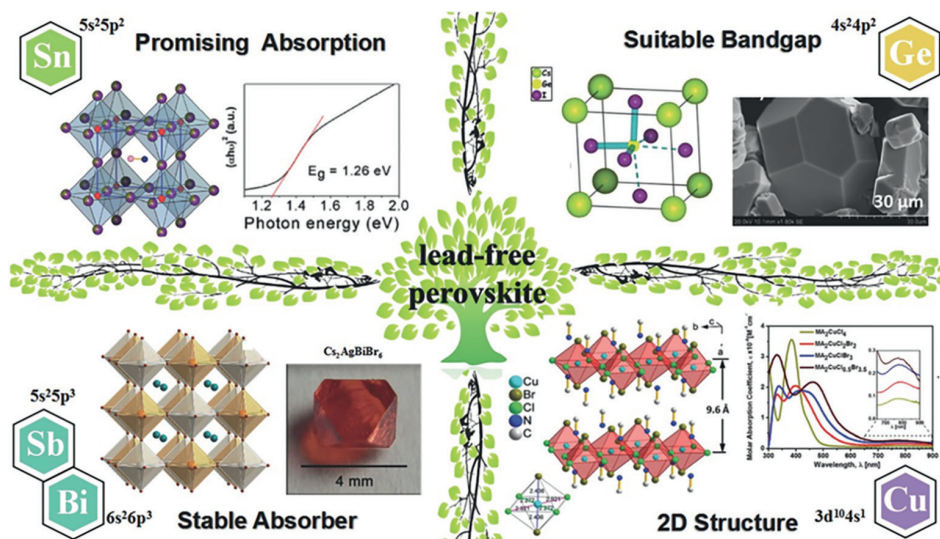


Figure 4. Several common organic–inorganic hybrid lead-free perovskite structure types [31].

of the cell is caused by the oxidation-induced generation of Ge^{4+} ions, even though MAGeI_3 has a band gap of 1.9 to 2 eV [33].

Researchers are interested in enhancing photovoltaic cells (PCE) with lead-free halide perovskite materials because of their high carrier mobility, electron transfer kinetics, and environmental friendliness. The right band gap, low background charge carrier density, low carrier effective mass, high absorption coefficient, and greater stability at the ambient atmosphere are essential factors for the champion device [34]. Up to now, tin-based halide perovskites have demonstrated a 14% power conversion efficiency in a single junction. Compared to single junction, hybrid tin and germanium halide perovskite exhibits higher power conversion efficiency and improved stability in ambient air [35]. Although the morphology of bismuth halide perovskite material is poor due to rapid crystallization, it has superior stability, which could affect device performance [36].

Numerous tactics, such as the addition of additives, reducing agents, 2D structure components, and solvent engineering device structure, are used to get past these obstacles. The shape of the film is primarily responsible for the lead-free halide perovskite's high power conversion efficiency. The choice of solvent, concentration of the solution, sample annealing, and technique (one-step spin coating, two-step spin coating, vapor-aided solution process, antisolvent processing) all have an impact on the film morphology. When it comes to the device's performance, the stability of the perovskite material has finally proven to be a major challenge. Halide perovskite materials based on tin and germanium quickly oxidize because of chemical instability in the surrounding atmosphere. Although bismuth-based halide perovskite material has a high stability compared to other perovskites, its rapid crystallization results in poor morphology. Thus, in order to build a lead-free halide perovskite solar cell with improved stability and high efficiency, these aspects must be taken into consideration.

These limitations can be solved thanks to the unique advantages of the newly developed solar cells based on hybrid organic–inorganic perovskites. The novel class of optoelectronic semiconductors referred to as organic–inorganic hybrid metal halide PSCs has garnered significant attention from photovoltaic researchers across the world. The advantages of hybrid organic–inorganic perovskites, which are the basis of the recently released solar cells, allow for the circumvention of these constraints [37]. An improved nanocomposite's mechanical properties result from the addition of additives near the interface. Their continually rising productivity, easy-to-use solution processing methods, usage of reasonably priced, lightweight materials, and other notable attributes are the reasons for this [22]. Hybrid perovskite research is still in its early stages; therefore, before PSCs can be used in practical applications, more fundamental research is needed to fully comprehend their electrical, optical, and physical characteristics.

2.3 Fabrications

In recent years, since PSCs' performance has greatly increased, silicon solar cells are no longer thought to be a better option. In this work, we have discussed a number of ways to improve the efficiency of perovskite solar cells, such as using different manufacturing processes, creating new materials for perovskite and charge transport, utilizing band gap engineering developments recently, and addressing stability concerns [38]. Despite much research, there are still numerous barriers to PSC improvement.

Moreover, most techniques for synthesizing perovskite materials, such as solvent diffusion, hot injection, inverse temperature, solvent evaporation crystallization, and anti-solvent vapor assisted, are based on the solution process [39]. One-step and two-step deposition techniques are among the solution synthesis methods for perovskite film, which combine all these constituents in an ordered network structure during manufacturing. With the sizes of the B- and X-site ions adhering to the octahedral factor limit, the $[BX_6]^{4-}$ octahedra composed of a core metal cation (B^{2+}) and halogen anions (X^-) create tetragonal or cubic network architectures *via* corner-sharing connections. Diverse perovskite formation processes result from variations in the unit cell's composition and characteristics, which in turn provide a range of formation energies [40]. The choice and optimization of a particular deposition technique are influenced by a number of variables, including the highlighted application's specific requirements, cost-effectiveness, and scalability.

Therefore, creating more synthetic paths to the solution process is crucial. To further optimize the physical properties of perovskite materials, morphologically low-dimensional structures, including nanorods, nanocrystals, nanoplatelets, nanosheets, and nanowires, can be synthesized [40, 41]. The process of trial and error is still largely used in the synthesis of novel perovskite materials. The cultivation of superior crystal structures with controlled dimensionality with no flaws requires the creation of a thorough design principle [42].

In recent years, there has been a growing interest in perovskite-based double junction tandem solar cells, such as perovskite/silicon, perovskite/perovskite, and perovskite/Cu(In, Ga)Se₂ [43], although the one-step deposition approach has been used to create the majority of wide-band-gap perovskite solar cells to date. Researchers have successfully built a perovskite solar cell with an active layer of 1.63 eV bandgap and a PCE of 20.35% utilizing the two-step sequential deposition approach [44]. By using the same material method as in the one-step method, the compositional optimization allowed for an improvement in FF to 81.53% and PCE to 20.35%, which was thought to be the highest efficiency of wide-band-gap fabricated by the sequential deposition method, when compared to the best device (an FF of 72.66% and a PCE of 17.82%) produced by the two-step method [45]. Wide bandgap perovskite materials, with their appropriate bandgap range of 1.60–1.72 eV, are good absorber materials to achieve high efficiency and current alignment.

Illustrated in **Figure 5**, the structure of a tandem solar cell consists of an n-type silicon bottom sub-cell and a lead-free perovskite top cell (hole transport layer: NiO; absorber: Cs₂AgBi_{0.75}Sb_{0.25}Br₆; electron transport layers: PCBM and SnO₂). A substantial open-circuit voltage of 1.76 V, a short-circuit current of 16.01 mA.cm⁻², and a PCE of 24.4% are attained by the improved tandem solar cell [46]. The benefits of the lead-free perovskite tandem solar cell, which combines acceptable thermal expansion behavior with electrical parameter values and efficiency that are very competitive with standard silicon cells, are also demonstrated by the results of a simulation of the effect of the working temperature. This kind of lead-free perovskite/silicon tandem solar cell is an excellent candidate for possible uses in optoelectronics and photovoltaics due to its non-toxicity and remarkable moisture stability. Because of this, perovskites are especially well suited for tandem use. In the world of photovoltaic energy, tandem solar cells that mix silicon and perovskites have been deemed the best options [47].

One of the simplest ways to reduce the levelized cost of electricity (LCOE) is to increase power conversion efficiency through the use of the tandem strategy. The sensitized structure, or tandem film structure, of each of these perovskite solar cells

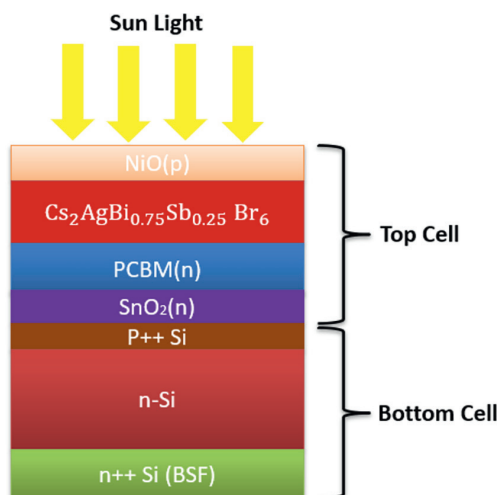


Figure 5. Structure of a tandem solar cell consisting of an n-type silicon bottom sub-cell and a lead-free perovskite top cell [46].

involves the formation of heterojunctions between the perovskite material and a variety of other materials. Typically, this approach entails stacking two sub-cells on top of one another and joining them using a 2-terminal (2 T) monolithic design or a 4-terminal (4 T) architecture [48]. The tandem concept has drawn a lot of interest because of its high open-circuit voltage (VOC) and overall power conversion efficiency. The most efficient tandem solar cells are built on the foundation of III-V semiconductor technology. They have achieved an efficiency of 32.8% for two-junction cells and 38.8% for five-junction cells in non-concentrated AM1.5 sunlight [49].

Perovskite/silicon tandem solar cells have advanced quickly as a result of advances in interface engineering, tandem structure design, and perovskite component optimization. Furthermore, it is essential to consider important elements while developing 2 T perovskite/silicon tandem devices, such as the top perovskite cell with a broad bandgap [49]. A significant barrier to the development of low-cost, high-efficiency tandem devices has been the lack of a high-efficiency top cell with a bandgap larger than 1.5 eV. Despite this, the high cost of materials and epitaxial layer development has limited the application of these tandem devices to specific industries like concentrator photovoltaics.

3. Conclusion

This review chapter highlights the necessity of a thorough design production that goes beyond the process of trial and error to direct the synthesis of perovskite materials. Additionally, it recognizes the early stage of hybrid perovskite research, highlighting the need for basic study to fully comprehend the material's electrical, optical, and physical characteristics prior to any practical uses, especially in perovskite solar cells (PSCs). The investigation of substitute synthetic paths for the widely employed solution technique is emphasized, recognizing the drawbacks and looking for creative solutions.

A variety of materials are introduced for usage as active layers, hole transport layers (HTL), and electron transport layers (ETL) as this chapter explores the

structure and tandem film structure of perovskite solar cells. Notably, the emphasis is on how important it is to create new, affordable hole transport materials in order to support perovskite solar cells. It is vital to create innovative, affordable hole transport materials to support the commercialization of perovskite solar cells. Furthermore, the industrial promotion and development of PSCs will be hampered by the exceedingly hazardous nature of the Pb element utilized in perovskite solar cells. Consequently, a low-toxicity or nontoxic component needs to be discovered in the future to replace Pb.

Furthermore, in terms of potential and practical industrial use, the monolithic two-terminal tandem cell is the best option. Si bottom cells that are sold commercially have an efficiency that approaches their theoretical maximum rather closely. The development of tandem devices is still hampered by a number of issues, such as scaling up manufacturing, the cost of acquisition, the inherent instability of perovskite materials, and performance improvement. The advantages of multifunctional cells have been highlighted due to the demonstration of intrinsic defects in single-junction solar cells. Due to the strengthening of tandem structures combined with perovskite/contact materials, efficiency records have been broken quickly. For the commercialization of perovskite/Si tandem technology, lab-scale 2 T perovskite/c-Si tandem efficiency has promise.

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