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Original Research Article

Perovskite–silicon tandem solar cells: A path to high efficiency

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ABSTRACT
Perovskite-based tandem solar cells (TSCs) have emerged as a promising technology for overcoming the efficiency limits of single-junction solar cells. Perovskite materials offer several advantages, including high absorption coefficients, tunable band gaps, long charge-carrier diffusion lengths, low-cost solution processability, and excellent compatibility with tandem solar cell architectures. Conventional crystalline silicon solar cells dominate the market, however, their power conversion efficiency is nearing the theoretical Shockley–Queisser limit, which significantly constrains further performance enhancements. Perovskite–silicon tandem architecture enables more efficient use of the solar spectrum, minimises thermalisation losses, and delivers substantially higher power conversion efficiencies than conventional single-junction solar cells. This paper provides a comprehensive analysis of perovskite–silicon tandem solar cells, with emphasis on their operating principles, device architectures, and material properties. While efficiency has improved considerably, several important challenges persist, such as maintaining long-term stability, mitigating environmental degradation, addressing lead toxicity concerns, and enabling scalable manufacturing.

1. Introduction

Over the past forty years, the exceeding global demand for energy has been met by fossil fuels. In 2018, the composition of the world's primary energy utilisation was as follows: oil constituted 33.6%, coal 27.2%, natural gas 23.9%, hydroelectric power 6.8%, nuclear energy 4.4%, and renewable energy sources 4% [1]. Fossil fuels accounted for 84.7% of the global primary energy utilisation during that year (BP 2019) [1]. This significant energy utilisation resulted in drastic climate change [2]. A photovoltaic (PV) solar cell is an electronic device that converts sunlight directly into electricity using the photovoltaic effect. According to the reported data, approximately 95% of the total market share of solar cells comes from crystalline silicon materials [3]. The main reasons for silicon's increasing popularity within the PV market are that silicon is available in abundance, and relatively much cheaper and sustainable compared to rare materials like Cd, In, or Ga. Silicon-based solar cells can be of two types: monocrystalline or multicrystalline, depending on the presence of one or many grains in the microstructure, which affects the solar cell properties, like their efficiency and performance. The current laboratory record efficiencies for monocrystalline and multicrystalline silicon solar cells are 26.7% and 24.4%, respectively [3].

Perovskite/silicon tandem-based solar cells can offer an effective path to increase the power conversion efficiency of crystalline silicon (c-Si) solar cells beyond the theoretical efficiency limits of single-junction cells. After their inaugural demonstration in 2015, perovskite/Si tandem solar cells have been able to achieve exceptional technological progression, reaching a certified power conversion efficiency of 34.9% by 2025 [4]. For the past decade, progress has been made in the direction of the fabrication of highly efficient laboratory-scale tandems using a range of vacuum and solution-based perovskite processing technologies utilising various types of c-Si bottom cells. However, to become industrially ready, the transition from laboratory to industrial fabrication will require appropriate, scalable, and stable input materials and manufacturing processes for the production of market-ready tandem cells. In addition, research on perovskite/silicon tandem devices needs to pay closer attention to scalability, stability, and reliability at every stage, from fabrication and characterisation to cell-to-module integration, along with better ways to predict and evaluate real-world performance [5].

2. Organic-inorganic hybrid perovskites

Perovskite solar cells (PSCs) contain perovskite materials, which are formed by hybrid organic/inorganic compounds of structural formula ABX_3, where X is a halide (I, Br, Cl). A is a monovalent cation part of the compound (Methylammonium, Formamidinium, or Caesium), and B is a divalent metal part of the compound (Lead or Tin). The active layer in perovskite material, which is deposited between two electrodes, (TCO) transparent conductive oxide and metal, consists of the two charge transport layers ETL and HTL. PSCs have many advantages, for example, a higher absorption coefficient, a longer diffusion length, and a slow rate of recombination.

Factors like these increase the power conversion efficiency (PCE), as they increase both open circuit voltage (V_{oc}) and short-circuit current density (J_{sc}). The standard PSCs devices consist of five main layers: conducting substrate indium tin oxide (ITO), the HTL, PV active material layer, ETL, and the metal electrode (Cu, Au, or Ag). After the solar cell is illuminated, the ETL/HTL extracts photogenerated electrons/ holes from the perovskite light-absorbing layer and then transports them to the cathode/ anode, as shown in Figure 1 [6]. The crystal structure of ABX_3 is significantly dependent on the reaction temperature and the ambient temperature [7].

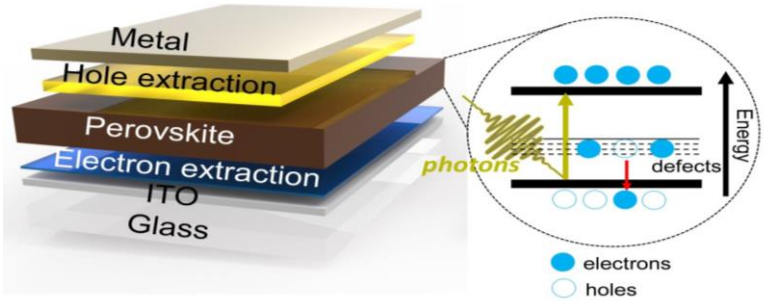

Figure 1: Schematic representing the construction of a perovskite solar cell [6]; Reproduced from © OIST (Okinawa Institute of Science and Technology Graduate University), licensed under the Creative Commons Attribution 4.0 International License (CC BY 4.0).

3. Working principle of tandem cells

Perovskite-silicon tandem solar cells operate differently compared to the single-junction solar cells, as their performance is constrained by the inherent optical bandgap. The tandem device combines semiconducting materials of different band gaps that are able to react with a wider range of the solar spectrum, generating power that is greater than the Shockley-Queisser (S-Q) limit. In Figure 2a, tandem configuration, the top solar cell that has a comparatively wide bandgap absorbs photons that have high energy (such as ultraviolet and visible light), while the bottom solar cell, which has a comparatively narrow band gap, absorbs photons that have low energy, near the infrared part of the solar spectrum [8]. This is one way to reduce losses due to thermalisation in single-junction silicon solar cell devices [9]. Commonly, tandem solar cells can be designed in two configurations, as illustrated in Figure 2b. In a monolithically integrated architecture, the wide-bandgap top cell is directly fabricated on the narrow-bandgap bottom cell, resulting in a series electrical connection between the two subcells, known as a two-terminal (2T) tandem device. Alternatively, in a mechanically stacked configuration, the subcells are optically coupled but remain electrically independent, forming a four-terminal (4T) tandem device [8].

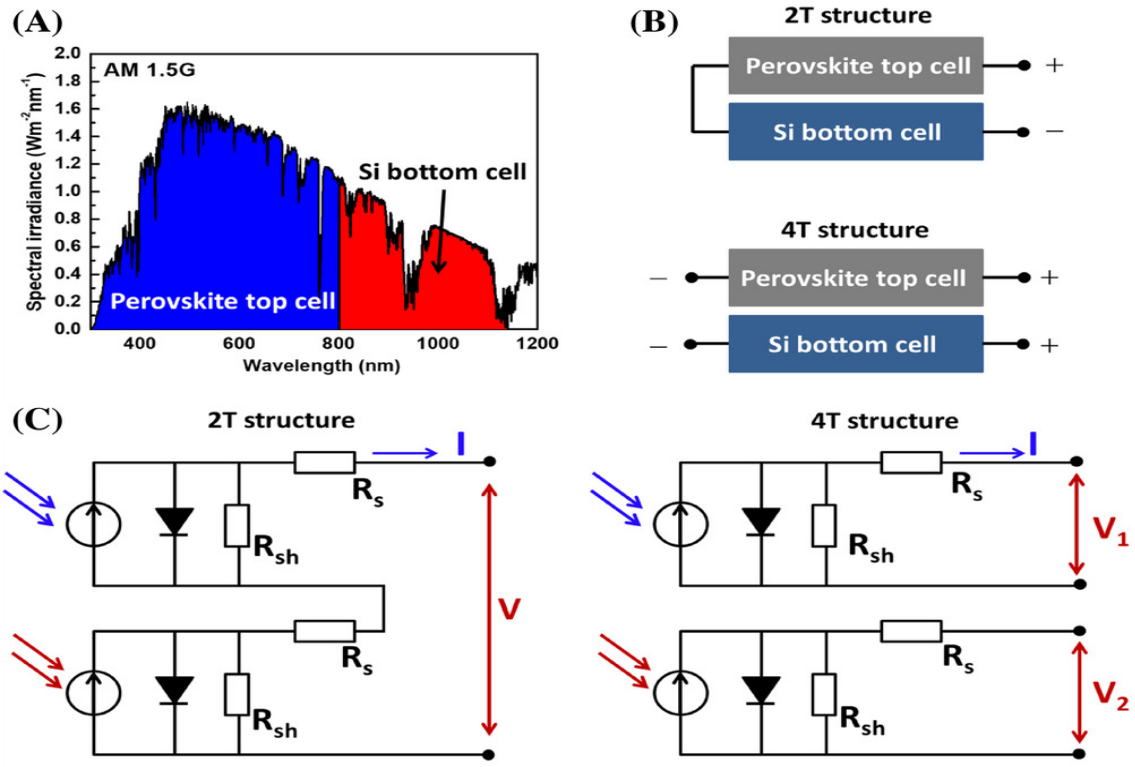

Figure 2: (a) Spectra response of the perovskite top cell and Si bottom cell in a tandem configuration. (b) Demonstration for the device structure of a 2T and 4T perovskite/Si tandem solar cell. (c) The equivalent circuit of a 2-terminal and 4-terminal perovskite/Si tandem solar cell under illumination. R_s is the series resistance, R_{sh} is the shunt resistance, I is the output current, and V is the output voltage [8]; Reproduced from Cheng and Ding (2021) under the terms of the Creative Commons Attribution 4.0 International License (CC BY 4.0).

In a 3T tandem cell, one more terminal is connected to extract current from PSC/Si [9]. Figure 2(c) shows us the equivalent circuit of perovskite/Si tandem solar cells. As the subcells in a 2T tandem device are connected in series, current matching of the top and bottom cells is always required to make sure the overall device current is not restricted by the subcell with lower current. In the case of 4T tandem solar cells, they do not need current matching because the top and bottom cells are electrically separated. Therefore, we need to either maximise the top cell or improve the efficiency of the bottom cell to boost the overall device performance of a 4T tandem.

In monolithic silicon−perovskite TSCs, different active layers are connected by interconnect layers (ICLs) to absorb light. For a short period, the wide band gap of the top PSC only absorbs high-energy photons and passes low-energy ones, which are absorbed by silicon with an energy band gap of ∼1.1 eV. Theoretical efficiency limits of PV/Si TSCS are constrained by silicon's band gap of ∼1.1 eV, and optimal pairing requires PSCs with an active layer of about ∼1.73 eV.

In 2T monolithic silicon−perovskite TSCs, generally, the top cells are of PSCs, and the bottom cells are silicon-based; both are connected through the ICLs. The top PSCs may be in the n−i−p or p−i−n structures, which have shown great results in silicon−perovskite TSCs. Nonetheless, PSCs with Spiro-OMeTAD as an HTL (hole transport layer) can produce the parasitic absorption of photons of high energy, hence limiting the J_{sc} in silicon−perovskite TSCs, which affects their performance. Parasitic absorption refers to light that is absorbed but does not contribute to the desired photocurrent generation. It is the light that gets absorbed by parts of the 2T silicon−perovskite TSCs other than the photoactive materials, limiting the overall PCE of the device. This can happen because of various components, such as contact layers, transparent conducting oxide (TCO), interconnecting layer (ICL), metal electrodes, encapsulation materials, and defects of active layer materials (perovskite and silicon) themselves. Basically, it restricts the amount of available light for the generation of photocurrent and hence limits the PCE. It can be reduced using TCOs, which have a wider band gap, along with lesser absorbance in the visible spectrum range of light [9].

4. Structure of tandem solar cells

4.1 Two-terminal monolithic tandems

The 2T tandem devices, consisting of top and bottom solar cells, are usually connected in series, which leads to an addition of the generated voltages and a recombination of the photogenerated currents of each subcell at the junction. Thus, to maximise the current generation of a 2T TSC, the photocurrent of the top and bottom solar cells needs to match. If this is not achieved, the total current produced by the tandem cell will be limited by the subcell that generates the lower current [10].

For the better performance of the sub-cells in a 2-T TSC, they need to have their photocurrents precisely matched. An interconnection layer (ICL) is used to connect the sub-cells in a 2-T TSC in series, making the device have properties of a single junction cell but with a larger power output. Usually, 2 T TSCs can also be measured using the same conventional techniques that are used in SJSCs. But we have a number of significant obstacles to accurately characterising 2-T TSCs, such as complicated electrical and optical interactions between the two sub-cells, spectrum mismatch, and difficulty in sub-cell characterisation [11].

In a 2T perovskite/c-Si TSC, the incident sunlight on the wide-bandgap perovskite results in absorbing the blue part of the spectrum while the 1.12 eV silicon absorbs the red part of the spectrum [12]. Simultaneously, the infrared photons that are passing through the top cell are then absorbed by the active layer of the narrow band gap bottom cell. The hole from the top cell and the electron from the bottom cell flow through the external load, resulting in the current. While electrons from the top cell and the hole from the bottom cell flow toward the ETL and HTLs of the cells, for the combination at the interface. But the issue arises when the ETL and HTL of two different subcells happen to be in contact, forming undesirable p−n junctions due to variations in their work function. These undesirable p−n junctions have energy barriers limiting the electron and hole recombination. This affects the V_{oc} and fill factor (FF) of the device. This is why monolithic silicon−perovskite TSCs require ICLs, which can be provided as layers for recombination or tunnelling. The ICLs deposited between the ETL and HTL are used to facilitate efficient charge recombination and are generally an entity of ETL, HTL, buffer layers, and recombination layers. ICLs are mainly based on TCOs (transparent conducting oxides), metal layers, and conducting polymers. The ICLs, of the tunnel type, use doping of n-type ETL and p-type HTL from different subcells, which form highly doped n^+−p^+ tunnel junctions. The TCOs, ITO (indium tin oxide), indium-doped zinc oxide (IZO), and zinc-doped tin oxide (ZTO) are the common ICL materials used in the fabrication of TSCs, because these materials allow the bottom cell to use a maximum spectral response and minimise their light parasitic absorption or reflection [9]. The solar spectrum can be utilised better, and the thermalisation loss of high-energy photons is decreased. The 2T perovskite/c-Si TSCs will be able to surpass the efficiency limit of single-junction silicon cells and PSCs [12].

4.2 Four-terminal tandems

A 4-T TSC can be constructed by stacking two solar cells as long as the top cell's back is transparent. Direct connection of two sub-cells is not necessary. The easy integration of 4-T TSCs makes them attractive, especially for solar cells, which have fabrication limits, even though there are possible optical losses and increased module prices due to the additional transparent electrode and insulation between the sub-cells [11].

The two subcells that make up this structure are arranged in a manner very similar to that of the 2T tandem solar cell. In contrast, although these cells are optically linked, they operate independently in terms of their electrical connections, as illustrated in Figure 3a. Each subcell can operate on its own and be adjusted separately because it has its own electrical contacts. This also gives more flexibility in choosing the bandgap of the top cell and makes the device less affected by changes in the light spectrum. Because of this, it has been observed that 4T tandem cells can attain higher levels of efficiency across a wide spectrum of bandgap values, which range from 1.6 to 2 eV for the upper subcell. When they are paired with a c–Si lower subcell, a higher value of 1.81 eV can be accomplished. In this design, the need for matching or interface layers between the two subcells is not very important. However, it requires the use of several transparent electrodes, which can cause unwanted light losses (parasitic absorption) [13].

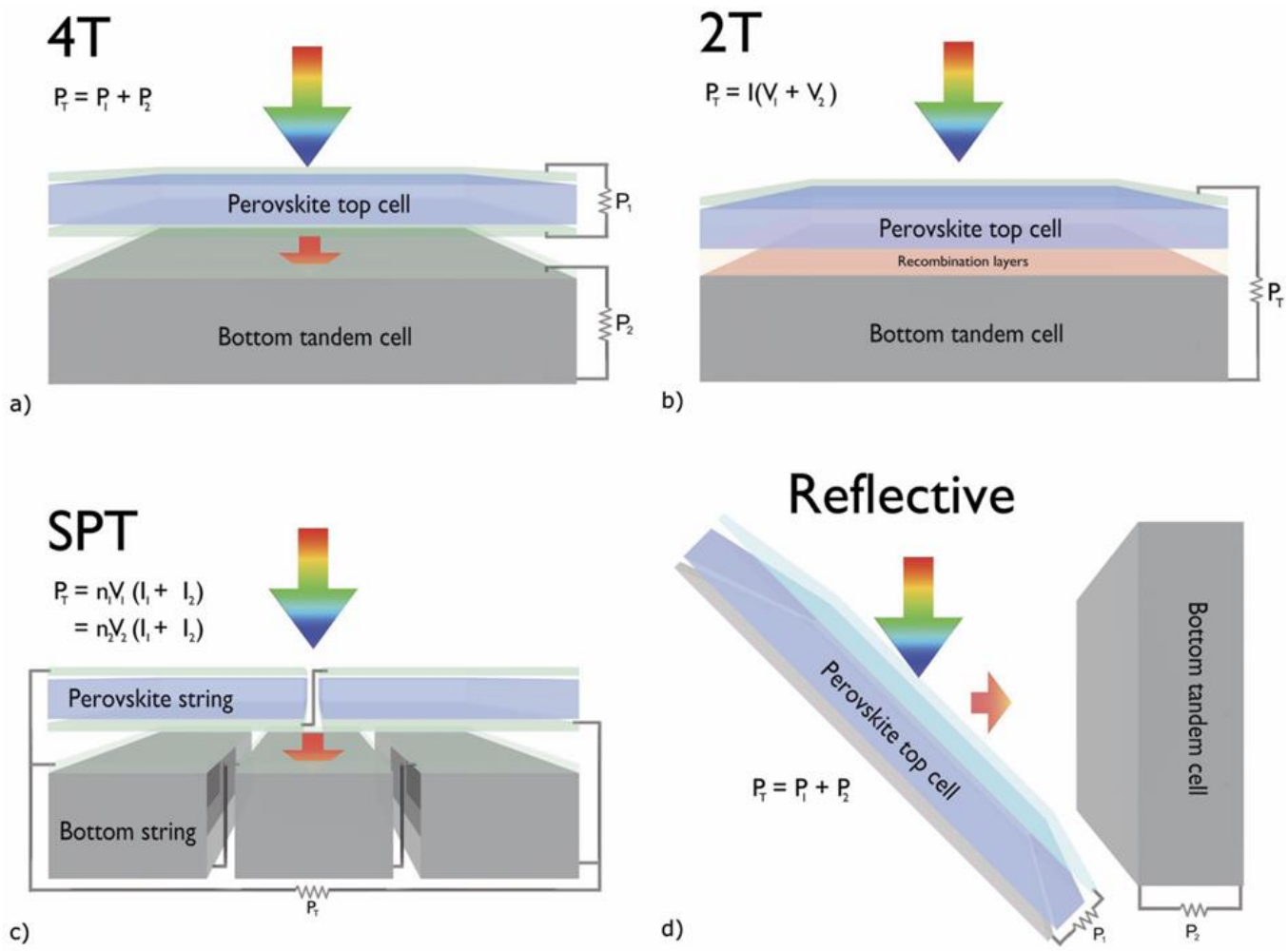

Figure-3: Varying degrees of electrical/optical independence in tandem configurations. (**a**) The 4T tandem structure has independent electrical connectivity to its two cells. (**b**) A 2T Tandem structure with series-connected elements. (**c**) Series-parallel tandem, voltage-matched sequences of top and bottom cells. (**d**) A reflective tandem with an infrared (IR) reflector, which is shown on the angled high-band gap cell [**15**]; Reproduced with permission from Lal et al., *Advanced Energy Materials* 2017, 7(18), 1602761. Copyright 2017 Wiley-VCH GmbH.

5. Efficiency of perovskite–silicon tandem solar cells

The theoretical efficiency limit of tandem solar cells under non-concentrated sunlight (AM1.5G spectrum) is 47% (Figure 4a), significantly higher than the Shockley-Queisser limit of 31% for single-junction cells under non-concentrated sunlight [15]. PV/Si tandem solar cells provide higher efficiencies than single-junction solar cells by absorbing higher-energy solar photons in a high-bandgap top cell material, where it can generate photocurrent with higher voltage than the underlying solar cell with a lower bandgap but broader absorption coefficient [15].

There are four main tandem solar cell configurations, as shown in Figure 4, each with varying degrees of optical and electrical independence. Four-terminal (4T) configurations (perovskite: c-Si) have achieved the highest perovskite tandem efficiencies of 25.2% to date [15]. Two-terminal (2-T) PV/Si tandem solar cells were made by fabricating perovskite cells directly on top of complete silicon bottom cells [16]. The monolithic perovskite tandem record is currently 23.6%, which has efficiency certified by the National Renewable Energy Laboratory (NREL), though the cell structure was not published at the time of publication [15].

Analysing both unconstrained 4T theoretical efficiencies given in Figure 4(a) and 2T tandems as shown in Figure 4b, using the results from Kurtz et al., employing a GaAs top-cell with optimised thickness for current-matching. Tandem solar cells 4-T and 2-T show a broad peak in the theoretical efficiency: 4-T tandem cells show a peak at 47% with a top/bottom cell bandgap pairing of 1.62/0.95 eV, and 2-T tandem cells at 39% with 1.72/1.14 eV [15]. But both configurations maintain high efficiencies within 10% of the peak for bandgap variations +/−0.1 eV on either side of the peak in both directions [15]. The decreased peak 2T efficiency of 39% compared to the 4T efficiency value of 47% is due to modelling of GaAs as the top-cell material of the 2T tandem, with non-ideal J_{sc}, though the trend for perovskite cells remains the same [15]. We now notice the particularly low sensitivity for 2T tandems when the bottom cell thickness is allowed to vary for current-matching (Figure 4b). Connection for submodules of two-terminal tandems (SPT) also affords greater flexibility in cell bandgap choice, with proper combinations matching the performance of four-terminal independently connected devices [15].

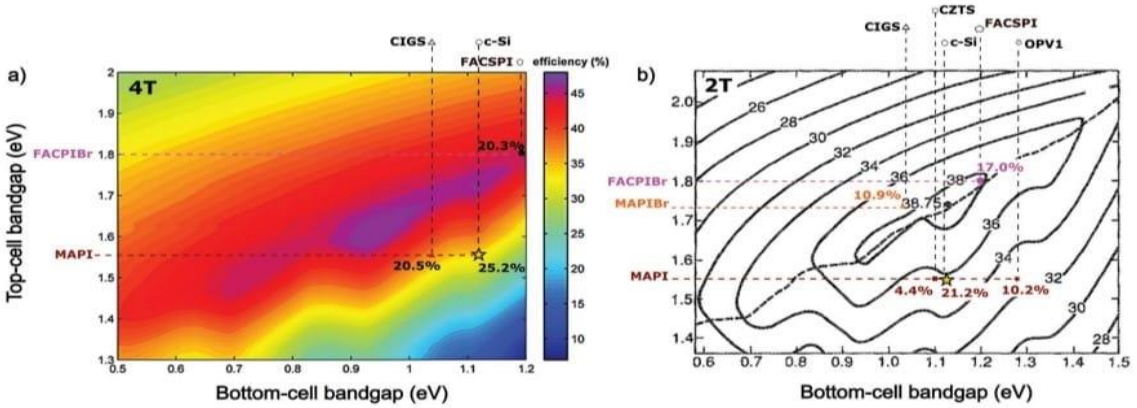

Figure 4: (a) Variation in the theoretical efficiency of four-terminal (4-T) tandem solar cells as a function of the absorber bandgap, together with the reported efficiency records of selected perovskite tandem solar cells [14, 15]; Reproduced with permission from Lal et al., *Advanced Energy Materials* 2017, 7(18), 1602761. Copyright 2017 Wiley-VCH GmbH. (b) Theoretical efficiency of two-terminal (2-T) tandem solar cells as a function of the bandgap for an optimized top-cell thickness, calculated using the absorptivity and dark current of a GaAs top cell, along with the reported efficiencies of series-connected perovskite tandem solar cells; Reproduced with permission from Lal et al., *Advanced Energy Materials* 2017, 7(18), 1602761. Copyright 2017 Wiley-VCH GmbH [15], which is adapted from Sarah R. Kurtz, P. Faine, and J. M. Olson, *Modeling of Two-Junction, Series-Connected Tandem Solar Cells Using Top-Cell Thickness as an Adjustable Parameter*, *Journal of Applied Physics* **1990**, *68*(4), with the permission of AIP Publishing [16]; Perovskite abbreviations used are as follows: $FA_{0.75}Cs_{0.25}Sn_{0.5}Pb_{0.5}I_3$ (FACSPI) and $FA_{0.83}Cs_{0.17}Pb(I_{0.5}Br_{0.5})_3$ (FACPIBr), $MAPbI_3$ (MAPI), $MAPb(I_xBr_{(1-x)})_3$ (MAPIBr).

6. Instability and degradation mechanisms in perovskites

6.1 Moisture and oxygen diffusion

The rapid rise in the efficiency of perovskite solar cells comes from better device designs, improved film quality, and smarter material choices. However, their long-term stability is still a concern, as materials like $MAPbI_3$ are quite sensitive to heat, moisture, light, oxygen, electric fields, and mechanical stress [18]. Many researchers have suggested that moisture induces reversible and irreversible degradation in perovskite films, as summed up in previous review articles. More importantly, water molecules can very simply enter into the perovskite structure, and it forms an intermediate monohydrate and dihydrate perovskite. The hydrate structures can completely form back to the original perovskite after 48 hours in dry air, which can be proven by X-ray diffraction. Water molecules, which are present in perovskite crystals, form very strong hydrogen bonds with organic cations, which undermines the bond between the cation and the PbI_6, which ensures faster deprotonation in the organic cation, and for external stressors, for example, heat or electric field, so this can be a better way for degrading the perovskite. Furthermore, water protonates iodide, creating volatile hydroiodic acid, HI. Due to this, yellow-coloured lead iodide (PbI_2) is obtained as a decomposition product [19].

Mechanism of degradation in the organic–inorganic halide perovskite was interpreted by referring to the X-ray diffraction (XRD) data before and after interaction with moisture.

$$CH_3NH_3PbI_3 \leftrightharpoons CH_3NH_3I(aq) + PbI_2(s) \quad \text{(i)}$$

$$CH_3NH_3I(aq) \leftrightharpoons CH_3NH_2(aq) + HI(aq) \quad \text{(ii)}$$

$$4HI(aq) + O_2 \rightleftharpoons 2I_2(s) + H_2O(aq) \quad \text{(iii)}$$

$$2HI \quad (UV) \rightleftharpoons H_2(g) + I_2(s) \quad \text{(iv)}$$

According to Niu and his team, the organic–inorganic perovskites go through decomposition in the presence of moisture, which causes the creation of CH_3NH_3I and PbI_2. Furthermore, CH_3NH_3I becomes CH_3NH_2 and HI in this process. Due to the oxidation process, HI becomes I_2 and H_2O. Simultaneously, a portion of HI undergoes photochemical reactions, due to which the creation of H_2 gas and I_2 occurs. The beginning of the degradation process is due to moisture; meanwhile, the decomposition rate is faster mainly because of oxidative (oxygen) and photochemical reactions (reactions caused by light).

Encapsulation is an effective strategy to protect devices by limiting the ingress of moisture and oxygen. While it does not entirely eliminate diffusion, it reduces it to negligible levels and helps resist the release of lead-containing degradation products. The encapsulant, which is easily available in the market for PSCs, is Ethylene Vinyl Acetate, polyisobutylene, etc [20].

6.2 Ion migration

Ion migration is one of the important factors that affects the working of PSCs. Ions (either anions or cations) inside halide perovskite materials are able to move under bias voltage or thermal drift, which leads to device uncertainty affecting the performance. Defects and ion migration, like the presence of iodine vacancies at the interface, are the reasons for degradation at the interface. Ion migration of defects affects the mechanism of operation in the device, which eventually will cause device failure when operating [20].

7. Sn, Bi-, and Ge-based lead alternatives for PSCs

The Pb is toxic in PSCs due to the presence of PbI_2 and $PbBr_2$, and if we compare, PbI_2 is more harmful than $PbBr_2$. If lead gets into the ecosystem it could also lead to serious damage to our wildlife and the environment. Studies have shown that one square meter of $MAPbI_3$ PSC modules on a rooftop can possibly decompose 0.9 g of PbI_2 because of intense rainfall.

In the past years, many studies have shown great interest in replacing the toxic lead from ABX_3 PSCs by using other elements, including tin, germanium, copper, antimony, or bismuth, although Pb is less costly. In all of the alternatives, Sn-based PSCs are convincing because of their better charge

mobility, long carrier lifetime, and smaller optical band gaps, which are quite close to the Shockley–Queisser limit. Attributes like these make them ideal and convenient to use for single-junction SCs and all types of perovskite-tandem SCs [21].

8. Future research directions

8.1 Interface engineering: use of 2D perovskites for passivation

Interface engineering using 2D perovskites is an important approach for surface passivation to improve efficiency and long-term stability. The goal is to use suitable passivating materials that interact well with the perovskite surface and help reduce defects and other barriers. Remarkably, the application of 2D perovskites and organic halides like phenethylammonium iodide (PEAI) has proved significant capability. Materials like these efficiently helped to reduce defects and provide reliability to the obstacles that are present at grain boundaries and on the surface of the perovskite layers. There is a layer formed that acts like a protective shield over the 3D structures of perovskite, which helps to reduce moisture and ion penetration and direct charge transport and recombination, which leads to better operational stability and efficiency of perovskite layers.

The existence of 2D perovskite protective layers helps to provide better optoelectronic properties for the device, which leads to enhanced and better light absorption and photoluminescence, advantageous to enhance the general operation of the PSC. For surface passivation when ammonium halides are used, a 2D perovskite layer will be formed on the outer surface of the 3D perovskite film. The general formula of these 2D layers is $(R\text{-}NH_3)_2PbX_4$; in this formula, R stands for an organic group, and X is a halide ion. The creation of 2-dimensional perovskite layers considerably influences the band structure of the PSC. These 2D layers usually contain bigger and larger band gaps compared to the 3D perovskite, which is under the 2D layer, forming a quantum well structure. This type of band alignment can be used for many reasons: acting as a barrier, to decrease the chances of carrier recombination at the surface, to help with efficient charge extraction, and to provide an additional protective layer to the underneath 3D perovskite, improving the overall performance of the device [22].

8.2 Flexible tandems

In the structure of flexible perovskite-based tandems, the top cell uses wide-bandgap near-infrared (NIR)-transparent PSCs, and for the bottom cell, the narrow-bandgap flexible PSCs. In the following two sections, we discuss the criteria and the recent progress for these two sub-cells. The main purpose of a flexible NIR-transparent perovskite as a top cell is to consume the solar energy, which is present in the UV and visible region, and these are designed to allow energy in the NIR region to pass through. Based on efficiency simulations, flexible NIR-transparent perovskite top cells should have a bandgap in the range of about 1.55 to 1.9 eV for 4-terminal (4-T) tandems. For 2-terminal (2-T) tandems, an optimal bandgap is typically around 1.8 eV (Figure 5). Materials like perovskite can serve many purposes, for example, tunable band gaps covering the target wavelength range, high absorption coefficient, sharp absorption edge and low temperature processability, and can be used as ideal absorbers for flexible top cells in tandem configurations. Being Efficient, flexible, and NIR-transparent are the three important factors of flexible NIR-transparent PSCs. Flexible NIR-transparent PSCs contain the structure of a flexible substrate/bottom electrode/electron transport layer (ETL) or hole transport layer (HTL)/perovskite/HTL or ETL/top electrode. The structure is dependent on the sequence of ETL and HTL; the devices can either be an n-i-p or p-i-n configuration [23].

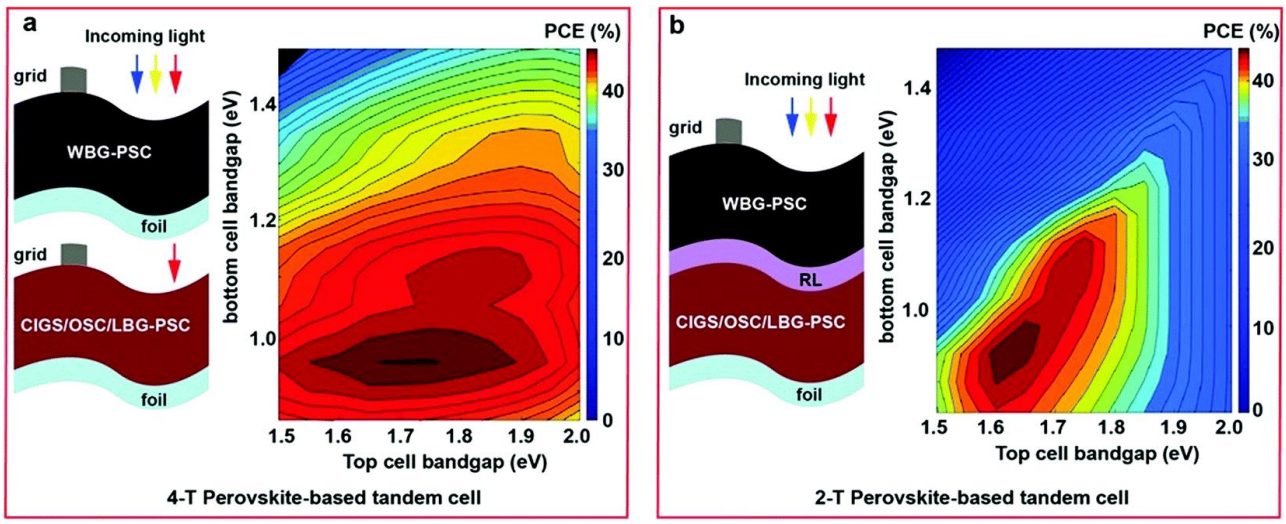

Figure 5: Illustration of the composition and efficiency of flexible tandem solar cells. (a) 4-Terminal tandem solar cells and (b) 2-Terminal tandem solar cells. Reproduced from Jiang and Qi (*Materials Chemistry Frontiers*, 2021), licensed under the Creative Commons Attribution-NonCommercial 4.0 International (CC BY-NC 4.0) License. [23], which is Reproduced from R. K. Kothandaraman *et al.* (*Small Methods*, 2020), licensed under the Creative Commons Attribution-NonCommercial (CC BY-NC) License [24].

9. Conclusion

Perovskite-based tandem solar cells (TSCs) have emerged as a promising technology for overcoming the efficiency limits of single-junction solar cells. By using a combination of materials with complementary band gaps, these devices will be able to provide better spectral utilisation and enhance PCE. Many advancements in device structure, material optimisation, and fabrication methods have shown that their development is for future photovoltaic applications. The significant challenges include long-term durability, moisture sensitivity, lead contamination, and commercial-scale processing. We can

overcome these issues by using material engineering, encapsulation methods, and the use of lead-free alternatives for practical applications. To summarise, the perovskite–silicon tandem technology represents a viable route towards high-efficiency and sustainable usage of solar energy, contingent upon ongoing research and development efforts to address current limitations.

Authors' contributions

All authors contributed equally to the conception, design, experimental work, data analysis, interpretation of results, and preparation of the manuscript. All authors reviewed and approved the final version of the manuscript for publication.

Conflicts of interest

The author declares no conflict of interest.

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Data availability

No new data were created.

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