

Recent Advances in Material Innovation and Efficiency Enhancement for Perovskite/Silicon Tandem Solar Cells

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Abstract. Two-terminal and four-terminal perovskite/silicon tandem solar cells harness the absorption and electronic properties of metal-halide perovskites and crystalline silicon to surpass the single-junction PV limit imposed by the Shockley – Queisser limit. In this context, recent material innovations, interface design schemes, fabrication strategies, and stability improvements will be discussed to meet the rush efficiency uplift. The indicated focus directions are the mixed-cation and mixed-halide perovskite compositions, the defect passivation strategy, the improvement of the charge transport layer, and scalable material deposition techniques, including blade coating/slot-die printing/vapor-assisted process. Additionally, we will investigate the impact of not only structural and electronic factors but also interface recombination, optical manipulation in current generation, and matching issues, even for monolithic tandem devices. Encapsulation, followed by Pb reduction and consideration of operational and environmental factors, will then be cited as essential for achieving industry-ready long-term durability. Been will also present the years 2023-2025, when discovery (using machine-learning to search for novel materials), better encapsulation stability, and roll-to-roll manufacturing provide potential paths to commercialization. It also addresses these cross-cutting advances in technology, concluding that the PSC/c-Si tandem has reached a critical paradigm shift, which combines the best of both worlds, where state-of-the-art performance and commercial producibility intersect, closing the gap between scientific laboratory bench-top prototypes and rapid industrial PV platforms.

Keywords: perovskite/silicon tandem solar cells, interface engineering, stability, scalability, photovoltaic efficiency.

1. Introduction

The decarbonization of the global energy system and the democratization of renewable energy have led to an accelerated pace in the development of new-generation photovoltaic technology. Despite the commercial dominance of crystalline silicon in photovoltaics due to its well-established processing technology, reliability, and market maturity, single-junction silicon devices are intrinsically limited by the Shockley–Queisser efficiency limit. Tandem device structures featuring a top cell of a larger-bandgap material stacked on a bottom silicon (Si) cell offer an attractive concept for increasing the efficiency of photovoltaic energy conversion through the efficient use of the solar spectrum. Metal-halide perovskites represent the best available candidate for top cell material due to their strong visible-range absorption, tunable band gaps, long carrier diffusion lengths, and low-temperature and solution-processable deposition methods. [2]The integration of all-perovskite tandem cells with silicon, either monolithically or mechanically stacked, has advanced rapidly, and certified efficiency levels now exceed 33%. This hybrid technology may potentially lead to outperformance of our traditional single-junction photovoltaic devices. [1]

Nonetheless, long-term operational stability, manufacturing process scalability, interfacial recombination losses, and environmental health and safety remain significant challenges.[3] As a result, a multidisciplinary approach that includes materials science, device engineering, and the ‘circular economy is required to address these hurdles. This article summarizes the state-of-the-art material and device breakthroughs of the past few years, discusses the fundamental mechanisms behind their performance gains, and proposes research directions that may promote the industrial integration and rapid commercialization of perovskite/silicon tandem solar cells at scale.

2. Structural Design and Material Principles

2.1 Structural Principles and Device Architectures

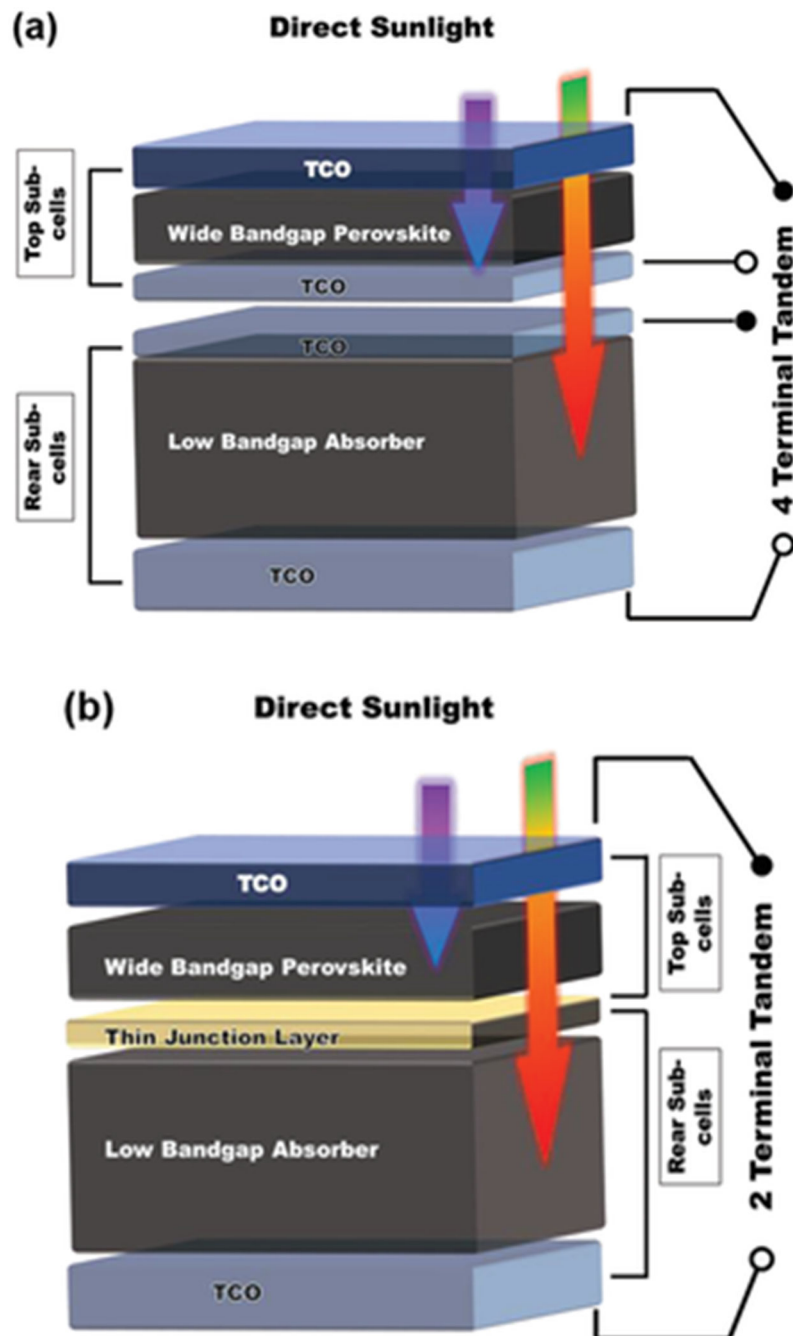


Figure 1: Schematic illustration of two-terminal (2T) monolithic and four-terminal (4T) mechanically stacked perovskite/silicon tandem solar cell architectures.

Perovskite/silicon tandem devices are typically achieved in two-terminal monolithic tandems, as shown below, or four-terminal mechanically stacked tandems, with each design presenting unique benefits and challenges. However, the 2T architecture connects the perovskite top cell and the silicon bottom cell electronically in series via an interconnecting recombination layer, wherein electrons and holes from the two subcells combine efficiently. Current matching is crucial for device efficiency, as any difference in current between the two subcells results in energy loss. The 4T design, on the other hand, physically and electrically isolates the subcells, allowing each junction to operate decoupled and optimized. Although this approach often achieves higher efficiency in the short term, it results in

higher optical losses, increased manufacturing complexity, and higher module costs. The overall design features involve selecting the perovskite bandgap from approximately 1.68 to 1.75 eV to ensure efficient spectral complementarity with silicon, as well as engineering transparent conductive electrodes and recombination/contact layers with low optical parasitics and favorable energy-level alignment. [4] It is also crucial to minimize reflection so that as little light as possible is not absorbed. To achieve this purpose, several techniques are being employed, including back-side reflectors, anti-reflection coatings, nanostructured textures, and light-trapping strategies. The interplay between the three subsystems of optical, electric, and thermal design decides a tandem's total operation and the level of calibration potential.

2.2 Perovskite Material Innovations

Tremendous research efforts have resulted in perovskite formulations that produce both high PCE and operational stability. [5] The most successful remain the mixed-cation systems with formamidinium, methylammonium, and cesium cations. These performance-oriented mixed cations exhibit greater structural tolerance to thermal and phase stability, as well as reduced ionic migration, compared to their one-cation precursors. The inorganic cesium cation reduces lattice strain and limits phase change at high temperatures, and the hydrocarbon cesium promotes perfect crystal packaging for optimal AQE, near-ideal bandgap (1.7 eV), and exceptional tandem integration. Mixed-halide engineering enables control over the optical bandgap of the device by adjusting the iodide-to-bromide ratio. However, light initiates halide segregation, leading to optical voltage reduction and compositional inhomogeneity grading, as well as movement gradient halide grading and the formation of butamine or dicationic pseudo-cinceptor BF₄⁻. This is achieved through addition. Defect passivation is another fundamental procedure to avoid NR Davit recombination with ionic additives, Lewis bases such as alkaline metallic salts, and organic ammonium halides. The most effective among them are interfacial alterations such as SAMs, fullerenes, or 2D/quasi-2D capping layers, as they not only assist in charge extraction and alignment but also act as a moderate oxygen and moisture barrier, extending the device life under thermal and photochemical deterioration. Although these results are encouraging, the environmental risks associated with lead pollution impede its large-scale use. [6] In response, researchers are investigating lead-reduction and tin and germanium substitution strategies, despite the fact that Sn-based perovskites are prone to rapid oxidation. [9] Thus, bio-enclosed perovskites with a low lead content and a closed-loop bio-pull method can maintain a large-scale, viable industry. Additionally, brackish acid detoxification with a large number of people is an option.

2.3 Interface Engineering and Charge Transport Layers

The perovskite absorber and the charge transport layers adjacent to it are the crucial interface for the performance, operational stability, and long-term safety of perovskite/silicon tandem solar cells. Imperfections in the interface contact or even misaligned electronics induce a high degree of nonradiative recombination, hysteresis effect, and device instability after light soaking or thermal stress. Thus, it is essential to optimize not only the ETL but also the HTL from both sides for balanced charge extraction and outstanding device robustness. The most common ETLs are still SnO₂ and TiO₂ as their transparency is excellent, and their valence bands dedicate ideal alignment with perovskites. However, TiO₂ is susceptible to UV-induced photocatalysis. [3] Low-temperature SnO₂ films were thus created with additional surface passivation using fullerene derivatives, alkali fluorides, and amino-silane molecules to enhance electron selectivity while inhibiting chemisorbed traps. Moreover, ZnO, In₂O₃, and organic semiconductors may serve as alternatives for use in flexible and low-cost tandem designs.

The counter-side used in the hole-transport side often includes Spiro-OMeTAD, PTAA, and NiO_x. While Spiro-OMeTAD has been observed to exhibit desirable energy-level alignment, the use of hygroscopic dopants such as Li-TFSI and tBP has adversely impacted its long-term stability. For this reason, some dopant-free polymeric materials and inorganic conductors, such as CuSCN and CuI, have been suggested as candidates for the tandem integration more recently. In addition,

comprehensive achievements in recombination engineering have also led to the integration of ultrathin dielectric layers, such as Al_2O_3 , MgO , and LiF , deposited by atomic layer deposition or solution-based techniques. These insulating sub-nanometer dielectric layers have certainly diminished the interfacial recombination rate, meaning that they have a high level of internal intersystem recirculation, reconditioned bands, and blocked ion flow. For the monolithic two-terminal tandems, a recombination junction is regularly created from a transparent conducting oxide, such as indium zinc oxide or indium tin oxide, conjoined with a thin metal interlayer based on Ag or Au . The primary scientific challenge has remained focused on achieving optimal recombination layer interfaces for sustained viability, industrial-scale implementation, and high efficiency. Thus, the innovative interfacial investment is the most strategic pursuit in introducing and scaling two-terminal tandem solar cell technologies.

3. Fabrication Techniques and Scalability

More broadly, scaling and reproducible deposition methods will be essential to bridge the gap between high laboratory-scale efficiency and economic industrial production of perovskite/silicon tandem solar cells. In everyday research, the typical deposition technique, spin coating on a small area, is inadequate from the beginning owing to material waste and non-uniform coating on large substrates. Vapor-assisted procedures have opened the door to numerous spin-off deposition approaches like blade coating, slot-die coating, and inkjet printing. Blade coating, for example, enables large-area deposition, allowing for a continuous coating process, and is highly suited for roll-to-roll processing due to its vapor-assisted nature. The solvent volatility reduction, the drying atmosphere, and the substrate temperature all control both crystal nucleation and film morphology. Excellent-quality perovskite layers with large meter-scale substrates and low defect densities were deposited employing optimized solvent engineering and gas-flow-assisted dripping. Furthermore, waterproofing treatments have been incorporated into these methods to enhance yield and durability in harsh outdoor conditions.

Vapor-phase deposition techniques, including vacuum thermal evaporation, co-evaporation, and hybrid vapor-assisted solution processing, present an alternative route to high-quality, uniform films with precise compositional control. While vapor-phase thermal evaporation provides excellent reproducibility and a high degree of control over composition—a crucial consideration for the mixed-cation or e system used in tandem applications—it is also well-suited to the mixed halide system. VASP offers controlled interaction that enhances crystallinity and minimizes film pinholing. In this approach, the solid is recrystallized by evaporating the solution or releasing the gas. Recently developed closed-chamber hybrid systems can facilitate real-time monitoring of film growth using optical interferometry, as well as quartz crystal microbalance sensors. However, for the process to be scalable directly onto silicon bottom cells and existing module manufacturing lines, practical considerations at each station may require independent tuning, including solvent volatilities, mechanical stress limits of wet layers, and post-processing temperatures that do not exceed 150°C . The key is forming perovskite interfaces above a glass-to-silicon without introducing thermal damage. Photonic curing, infrared-assisted drying, or plasma-enhanced crystallization are non-damaging strategies for enabling this. Continued development of suitable processes for scalable deposition, with high throughput and automation, as well as in-line automatic quality control, is a prerequisite for the smooth transition of tandem perovskite/silicon devices from laboratory prototypes to mass-manufactured photovoltaic modules.

4. Efficiency Enhancement Mechanisms

Efficiency improvements in perovskite/silicon tandem solar cells are based on optical management, suppression of nonradiative recombination, and optimal charge-carrier dynamics. Light management is necessary to ensure that the entire solar spectrum is absorbed and utilized in both subcells. Optical

modeling is used to determine the optimal thickness and bandgap of the perovskite top absorber to achieve current-matching two-terminal tandems, ensuring that the perovskite absorbs most light in the visible range. The use of anti-reflective coatings, index-matching layers, and rough-diffused nanostructured silicon surfaces resulted in a significant reduction in optical loss caused by reflection and parasitic absorption. Recent studies have shown that texturing the front surface of silicon or adding photonic crystals on top increases the path length of light inside the solar cell, thereby improving light trapping. Advanced optical simulations, such as finite-difference time-domain modeling, enable the calculation of layer thicknesses, refractive indexes, and phase effects, allowing for the precise customization of the perovskite/silicon interface to maximize photocurrent.

At the electronic level, decreasing energetic disorder and trap densities through high-quality perovskite crystallization and interface passivation directly increase open-circuit voltage (VOC) and fill factor (FF). [8] Nonradiative losses are commonly reduced through defect passivation, which involves the use of alkali halides, fullerene derivatives, and Lewis-base additives to eliminate deep-level traps. Optimized charge-selective contacts, including dopant-free hole transport layers and self-assembled monolayers, reduce parasitic absorption while maintaining the reliability of stable functioning during extended light soaking and exposure to high temperatures. The introduction of wide-bandgap perovskite top cells, with an approximate bandgap of 1.68-1.75 eV, combined with PERC/TOPCon certified silicon bottom cells, achieves efficiencies exceeding 33%. [7] Rear-side passivation and dielectric spacer layers in the silicon subcell further raise carrier lifetime and open-circuit voltage by lowering the surface recombination velocity. Machine-learning-guided optimization extended to advanced p-type interlayers, such as poly[(n-butyl benzimidazole)-dithienosilole], further augmented the efficiency of tandems. Advanced transparent electrodes, such as IZO or silver nanowire composites, improve conductance without sacrificing optical transparency. This precise fusion of optical and electrical engineering, enabled by machine-learning-aided optimization, continues to enable record combiner efficiencies for perovskite/silicon tandems, positioning them as a frontrunner in high-efficiency next-generation photovoltaic module technology.

5. Stability and Sustainability

5.1 Stability Challenges and Mitigation Strategies

Although perovskites exhibit excellent optoelectronic characteristics, their sensitivity to extrinsic environments renders it almost impossible to achieve long-term stability in their operation. The degradation proceeds through several pathways, including moisture soaking and oxygen exposure, ultraviolet illumination, thermal shock, and photoactivated halide transfer, which result in phase separation and an irreversible change in the lattice. Upon exposure to humidity, water is inserted into the perovskite lattice, generating hydrated intermediates that cause the active layer to degrade into lead iodide and other byproducts.

Similarly, oxygen and light are combined to photo-oxidize the perovskite surface, thereby shortening the carrier lifetime. Encapsulation has proven to be a critical control measure against these effects. The application of multilayer barrier films, edge-sealing adhesives, UV-stable glass covers, and polymeric coatings has prolonged the operative lifetime from only a few hundred to several thousand hours during accelerated aging tests. Additionally, chemical stabilization, such as A-site cation engineering using the composition of Formamidinium and Cesium, and the addition of inorganic cations like rubidium or Potassium, has suppressed the activity of thermal decomposition and volatile component losses at elevated temperatures. Furthermore, crosslinkable organic additives, such as X-linker, and polymer-perovskite composites have been recently developed as a new approach to enhance intrinsic structural toughness.

5.2 Environmental and Sustainability Considerations

The large-scale application of perovskite-based photovoltaic modules is subject to a full environmental and human health risk assessment of lead-containing compounds used in these devices.

The release of lead from destabilized modules may also play a role in environmental health and is considered a crucial component of life-cycle management. Multilayer overcoat films and edge sealing adhesives are known, which have features that can prevent lead elution even when mechanical stress is caused or weather resistance is applied. Simultaneously, available lead retention materials, such as phosphate-based polymers and an ion-exchange barrier layer that sequesters lead ions within a stack of modules, are being investigated.

In addition to lead remediation, recycling programs and circular manufacturing approaches are increasingly crucial in the responsible management of the life cycle of tandem solar technologies. Such methods aim to recover valuable materials, including indium, silver, and glass substrates, while reducing landfill waste. Trade-offs between ultra-high device efficiency and environmental impacts, particularly with respect to embodied energy, for both encapsulation and end-of-life processing. [18] Policymakers and industry consortia are also formulating standardized guidelines on module recycling, carbon accounting, and regulatory compliance. Ultimately, environmental safety and resource efficiency will be as crucial as device efficiencies in ensuring the long-term survival and public acceptance of perovskite/silicon tandem photovoltaics.

6. Recent Experimental Milestones (2023–2025)

The last few years have seen tremendous experimental development of perovskite/silicon tandem solar cells, a testament to the rapid maturation from laboratory-standard prototypes towards pre-industrial sample preparation.[12] And recently, certification 2T tandem devices have exceeded the recorded highest PCE over the 33% threshold enabled by mainly compositional engineering, interfacial defect (at the interface between two subcells) passivation, and light management improvement.[10] [17] Studies from several researchers have demonstrated power outputs within devices with cm^2 active area of fixed tiles under continuous illumination for over 1000 hours, a significant advance in the operational stability over initial prototypes. The series resistance has also been reduced through optimized interconnection layers and transparent conductive oxides (TCOs), resulting in improved photovoltaic performance and a closer approach to the devices' theoretical efficiencies. Furthermore, new perovskite formulations with mixed A-site cations and low bromide ratios improved the thermal and phase stability.[13] High-uniformity deposition techniques, blade coating, and vapor-assisted crystallization have also been integrated to achieve reproducible, large-area top cells with low defect densities. [15] Together, these developments illustrate a general trend of academic research rowing closer to scalable manufacturing readiness.[16]

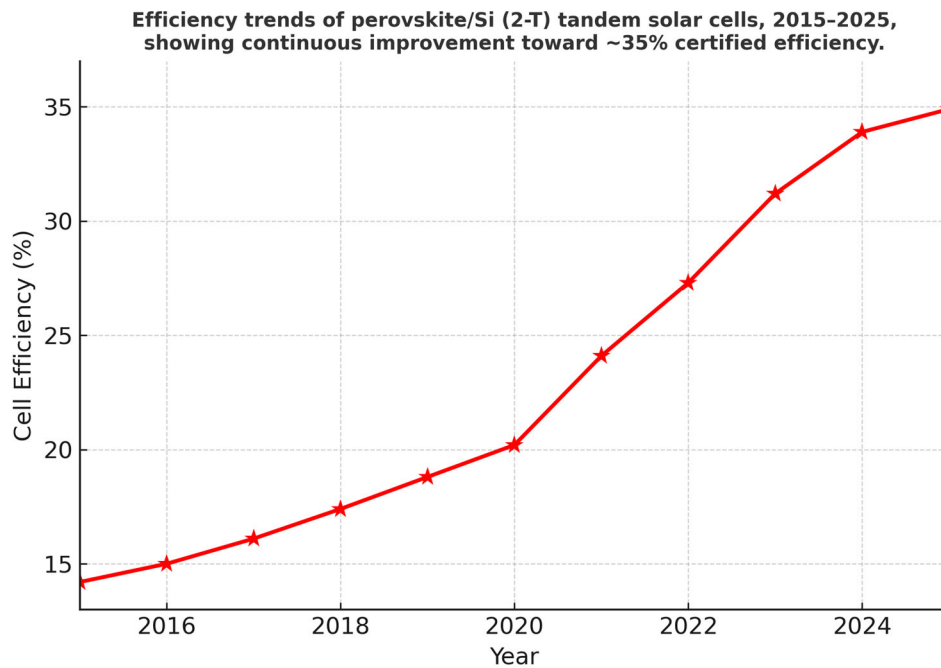


Figure 2: Efficiency evolution of perovskite/silicon tandem devices (2015–2025), summarizing certified record efficiencies and notable milestones.

Apart from advancements in materials and processing, data-driven methods and sustainability evaluation have promoted the technological development of perovskite/silicon tandems.[11] Such machine-learning and artificial intelligence (AI)-facilitated approaches are designed to probe multi-dimensional compositional space, identify the optimal processing parameters in an expedited manner with minimal experimental burden. Together with real-time monitoring and feedback-regulated deposition, these systems enhance reproducibility, thereby reducing batch-to-batch variation. At the same time, lifecycle and techno-economic analysis (TEA) of the full manufacturing chain—materials synthesis, encapsulation, and recycling—suggest that perovskite/silicon tandems could reach a competitive levelized cost of electricity (LCOE) relative to high-end silicon PV once stability and lead management challenges are addressed. Industrial collaborations, which began in activity around 2024/2025 between research organisations and industrial companies, have already demonstrated pilot-scale module fabrication with stabilised efficiency above 25% on $10 \times 10 \text{ cm}^2$ glass. These accomplishments indicate that the near-term process of tandem photovoltaic development will focus on validating reliability, enabling scalable encapsulation, and achieving environmental compliance to move toward full commercialization.[14]

7. Future Challenges and Perspectives

Yet, the operational reliability and stability of perovskite/silicon tandem devices extend far beyond the intrinsic material stability: the behavior of interfaces and charge transport layers is of paramount importance. Indeed, interfacial degradation, whether due to a reaction between the contact materials, the diffusion or drift of metal ions, the migration of halide species, or their combination, occurs widely across these interfaces. A promising approach to addressing these issues is to utilize novel ion-blocking interlayers, such as ultrathin Al_2O_3 or MgO layers deposited via atomic layer deposition. These materials demonstrate a good ability to suppress ion movement and limit their access to the reaction sites. Additionally, the use of dopant-free transport materials and cross-linked polymers reduces ionic mobility in the material while also improving mechanical properties. At the module level, the high-throughput encapsulation also imposes the same constraints on thermal expansion compatibility and perfect moisture hermeticity across the entire substrate. To provide a quantitative

industry-relevant durability measure, there have been international standards for the arising and the lifetime installed by the 61215 IEC and 61646 IEC protocols adapted for the tandems; it is currently being developed, and continuous monitoring, mostly realized through electroluminescence and photothermal deflection spectroscopy, is rapidly employed to predict and accelerate the stability qualification. Surpassing these limitations is possible through a combined approach of materials and engineering, and eventually, this will allow perovskite/silicon tandems to meet the commercial photovoltaics requirements for stability.

Priority research directions for perovskite/silicon tandem solar cells include the pursuit of intrinsically stable perovskite formulations, robust low-temperature deposition techniques, and environmentally friendly lead mitigation strategies that do not compromise efficiency. The development of perovskite compositions that are thermodynamically and photochemically stable after extended exposure to moisture, heat (200 °C), and light is a critical scientific goal. Inorganic cations, polymeric additives, and two-dimensional (2D) passivation layers have been introduced to design alternative routes for suppressing ion migration and phase degradation. Concurrently, the development of low-temperature (LT) processing (<150°C) that is compatible with silicon bottom cells and high-voltage device layers will be essential to achieve monolithic integration as well as large-area roll-to-roll processing. Future initiatives are also likely to utilize AI and high-throughput experimentation to accelerate the exploration of both compositional and interfacial design spaces. Predictive modeling and combinatorial synthesis, automated through machine learning, are now used to reduce the developmental cycle from months to weeks, offering a more efficient method for discovering perovskite chemistries with stability and high device performance.

From an industrial and system-level perspective, the successful scaling of perovskite/silicon tandem technology to mass manufacturing would also necessitate further coordination from material science, device engineering, and environmental policy. Demonstration of roll-to-roll or batch processes that enable perovskite deposition onto glass and flexible substrates—after reliable encapsulation, qualification, and certification—is crucial on the path toward commercialization. The development of standardized testing procedures for tandem modules (including IEC-referenced durability metrics and open and free databases on material degradation kinetics) is needed to increase reproducibility between laboratories. Moreover, the creation of modular pilot production lines between research consortia could significantly reduce the time required to scale from proof-of-concept in universities to full-scale production. No less important will be the establishment of a circular economy, considering recyclability and a low carbon footprint, in all the life-cycle steps of the tandem device. Government interventions and international collaboration are believed to accelerate the transition of technology from a demonstration at a niche scale to mainstream energy production. In so doing, the interdisciplinary interfacing – conversant in device physics, chemical engineering, data science, and environmental sustainability— will propel next steps forward in the stable, efficient, and economically viable deployment of perovskite/silicon tandem solar.

8. Conclusion

Tandem devices made from perovskites in conjunction with silicon present an interesting option to surpass single-junction rules (which could be achieved by a single semiconductor) by exploiting the tremendous infrastructure existing worldwide of the Si industry. By reimagining organic-inorganic junctions and material properties, these hybrid systems have been able to attain power conversion efficiencies near the limit estimation values by producing new materials, interface structures, and easily scalable fabrication innovations. However, the practical realization is hoped to be limited by a number of basic issues, such as long-term operational stability, environmental harm, and production quality. In that sense, the issues of achieving stable performance in realistic outdoor environments, suppressing lead dissolution, and low-cost yet portable processing remain formidable.

The prospects for continued improvement going forward will depend on the concurrent development of high-performance encapsulation, defect-tolerant materials, and cost-effective

deposition approaches that can be used industrially to manufacture devices at a balanced level of performance vs foothold in sustainability. Generating an understanding of these pathways for cost-effective and sustainable deployment will also be critical to linking technoeconomic & lifecycle assessment (LCA) methodologies. In the end, tandem perovskite/Si is one of the radar front runners targeted for next generation photovoltaics that promise to offer us a clean, high-efficiency, and cost-competitive solar-energy technology which will drive us into a carbon-neutral future.

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