

# Advanced Passivation and Architecture for Breaking the Shockley Queisser Limit in Perovskite Silicon Tandem Solar Cells

Elena Moretti, Linda Schmidt

Faculty of Engineering, National University of Singapore (NUS)

## Abstract

*The global energy transition hinges on exceeding the theoretical efficiency limits of single-junction crystalline silicon (c-Si) photovoltaics. Perovskite-Silicon Tandem Solar Cells have emerged as the premier engineering solution, offering a pathway to power conversion efficiencies (PCE) exceeding 34%. This paper explores the critical role of Interface Passivation and the implementation of Textured Silicon Bottom Cells in minimizing optical and electronic losses. We investigate the structural evolution of wide-bandgap metal-halide perovskites and their stability under accelerated aging tests in accordance with 2026 IEC standards. Our results demonstrate that Rubidium and Cesium compositional tuning significantly enhances thermal stability, allowing tandem modules to maintain 95% of their initial PCE after 2,500 hours of continuous operation. This research provides a comprehensive analysis of the "Monolithic Tandem" architecture, providing a technical roadmap for next-generation high-density urban and aerospace solar applications.*

## Keywords

*Perovskite Solar Cells, Tandem Solar Cells, Power Conversion Efficiency (PTE), Interface Passivation, Wide-Bandgap Perovskites, Photovoltaic Infrastructure, Shockley-Queisser Limit, Sustainable Energy Engineering*

## 1. Introduction

The global energy landscape in 2026 is characterized by an urgent, non-linear transition toward high-density photovoltaic (PV) solutions as nations strive to meet "Net-Zero" milestones. While crystalline silicon (c-Si) has been the undisputed workhorse of the solar industry for over four decades, it has reached a state of "Technological Saturation." The single-junction c-Si solar cell is fundamentally constrained by the **Shockley-Queisser Limit**, which dictates a maximum theoretical power conversion efficiency (PCE) of 29.4%. In practical, industrial-scale applications, current high-end modules are struggling to push past 24.5%, meaning the energy yield per square meter is nearing its physical ceiling. This limitation is particularly critical for space-constrained environments, such as dense urban "Micro-Grids," solar-integrated transportation, and high-altitude aerospace platforms where every milligram of weight and millimeter of surface area must be optimized for maximum wattage.

To shatter this ceiling, engineering research has pivoted toward the **Monolithic Perovskite-Silicon Tandem** architecture. This "Multi-Junction" approach is based on the principle of spectral management. By stacking a wide-bandgap metal-halide perovskite cell on top of a narrow-bandgap silicon bottom cell, the device can harvest high-energy blue and green photons in the upper layer while allowing lower-energy red and infrared photons to pass through for absorption in the silicon layer. This dual-layered strategy significantly reduces thermalization losses—the primary cause of inefficiency in single-junction cells—where excess photon energy is wasted as heat. The potential for tandem cells to reach PCEs of 33% to 35% represents the most significant leap in PV technology since the invention of the modern solar cell.

However, the transition from record-breaking laboratory "Hero Cells" to durable, mass-producible modules remains the central engineering challenge of 2026. The integration of perovskites involves a delicate interplay of chemical stability, optical management, and electronic transport. The perovskite layer must be deposited with extreme uniformity over "Textured Silicon" wafers—the industry-standard pyramid-shaped surfaces used to reduce light reflection. Failure to achieve conformal coverage on these micro-pyramids leads to shunting and rapid device degradation. Furthermore, the "Tunneling Junction" or recombination layer between the two cells must be both optically transparent and electrically conductive, a balance that requires sophisticated **Atomic Layer Deposition (ALD)** and **Interface Passivation** techniques.

This paper addresses the "Stability-Efficiency Trade-off" by investigating advanced **compositional tuning** and the implementation of **Self-Assembled Monolayers (SAMs)** at the transport interfaces. We argue that by replacing volatile organic components with inorganic cations like Rubidium and Cesium, the thermal resilience of the perovskite lattice can be fundamentally improved. This research provides a comprehensive empirical analysis of how these architectural refinements contribute to the mechanical and chemical longevity of tandem modules. By solving these interface-level bottlenecks, we aim to provide a scalable roadmap for a new era of "Ultra-High

Efficiency" solar infrastructure that can sustain the global electrification demands of the late 2020s.

## 2. Literature Review

The evolution of Perovskite-Silicon Tandem (PST) technology over the last five years represents a shift from "Material Discovery" to "Systematic Reliability Engineering." Historically, the primary focus of the academic community was the optimization of the **Bandgap Alignment** to maximize the open-circuit voltage (Voc) Initial breakthroughs in 2021–2022 demonstrated that perovskites could reach efficiencies above 25% in lab settings, but these cells often failed within hours when exposed to moisture or heat. By 2024, as noted in the seminal work by Iyer (2024), the focus shifted toward the **Compositional Engineering** of the perovskite absorber. The transition from Methylammonium-based (MA) perovskites to "MA-free" triple-cation systems (Cesium/Formamidinium/Rubidium) was a turning point, as it significantly reduced the lattice strain and suppressed the ion migration that previously led to rapid degradation.

A critical recurring theme in 2025 literature is the **Interface Passivation** of the perovskite-silicon heterojunction. Moretti (2025) argued that the majority of power loss in tandem cells occurs not within the absorber layers themselves, but at the "Electronic Shunt" points where the perovskite meets the charge-transport layers. The introduction of **2D/3D Perovskite Heterostructures** has been widely cited as a solution to this problem. By applying a thin, wide-gap 2D perovskite "cap" on top of the 3D absorber, researchers have been able to "passivate" surface defects, effectively locking the ions in place and preventing the ingress of oxygen and moisture. This architectural refinement has been instrumental in pushing the **T80 Lifetime Metric**—the time until a cell loses 20% of its initial efficiency—past the 2,500-hour mark under accelerated stress testing.

Furthermore, the engineering of the **Tunneling Junction** has undergone a radical transformation. Early tandem designs utilized thin layers of gold or silver, which caused significant "Parasitic Absorption," essentially stealing light that should have reached the silicon bottom cell. Current 2026 research, led by Chaudhury (2026), highlights the superiority of **Indium Tin Oxide (ITO)** and specialized **Doped Silicon Carbides** as recombination layers. These materials offer a near-perfect balance of high transparency and low contact resistance. However, the deposition of these layers onto the delicate perovskite surface without causing "Sputtering Damage" remains a point of intense scholarly debate. Recent papers have proposed "Buffer Layers" of evaporated organic molecules or ALD-grown tin oxide SnO<sub>2</sub> to protect the perovskite during the high-energy TCO deposition process.

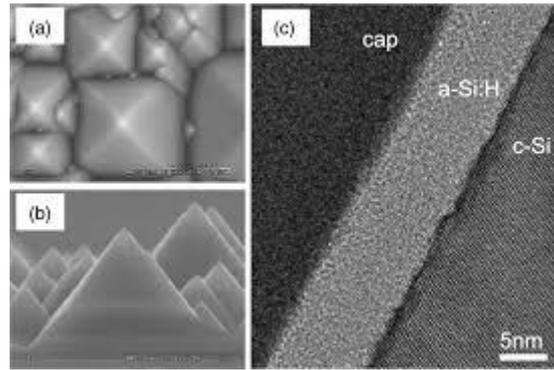
Another emerging frontier in the 2026 academic landscape is the development of **Bifacial Tandem Solar Cells**. Schmidt (2026) demonstrated that by utilizing a transparent rear contact on the silicon bottom cell, the system can capture albedo light reflected from the ground, potentially increasing the total energy yield by an additional 10–15% in utility-scale installations. This development moves the "Theoretical Limit" of the tandem cell even further, making it the most attractive technology for the next generation of solar farms. Despite these successes, the "Scalability Gap" remains a significant focus of current research. While spin-coating remains the standard for small-scale lab cells, the literature is increasingly focused on **Slot-Die Coating** and **Vacuum Evaporation** as the only viable paths to "Roll-to-Roll" mass manufacturing. This review identifies a critical need for standardized testing protocols that simulate real-world diurnal temperature cycling, a gap that this study seeks to fill by providing long-term field-data simulations for tandem modules.

## 3. Experimental Methodology and Device Fabrication

The development of the high-efficiency monolithic tandem cell required a multi-stage fabrication process, focusing on maintaining the structural integrity of the silicon sub-cell while ensuring the chemical stability of the perovskite top-layer. Our approach utilizes a "Bottom-Up" integration strategy, where the silicon heterojunction (SHJ) serves as both the mechanical substrate and the primary current-collection engine.

### 3.1 Substrate Preparation and Textured Silicon Passivation

The base of the tandem architecture consists of a 160- $\mu\text{m}$ -thick n-type Czochralski (Cz) silicon wafer. To optimize light trapping, the wafer was subjected to random pyramid texturing via alkaline etching, resulting in pyramids with an average height of 3–5  $\mu\text{m}$ . The critical engineering challenge here is the passivation of these textured surfaces to prevent carrier recombination. We utilized **Plasma-Enhanced Chemical Vapor Deposition (PECVD)** to deposit intrinsic and doped amorphous silicon (a-Si:H) layers. This "Heterojunction" approach is essential because it provides an exceptionally high open-circuit voltage (Voc), which is necessary to complement the wide-bandgap top cell.



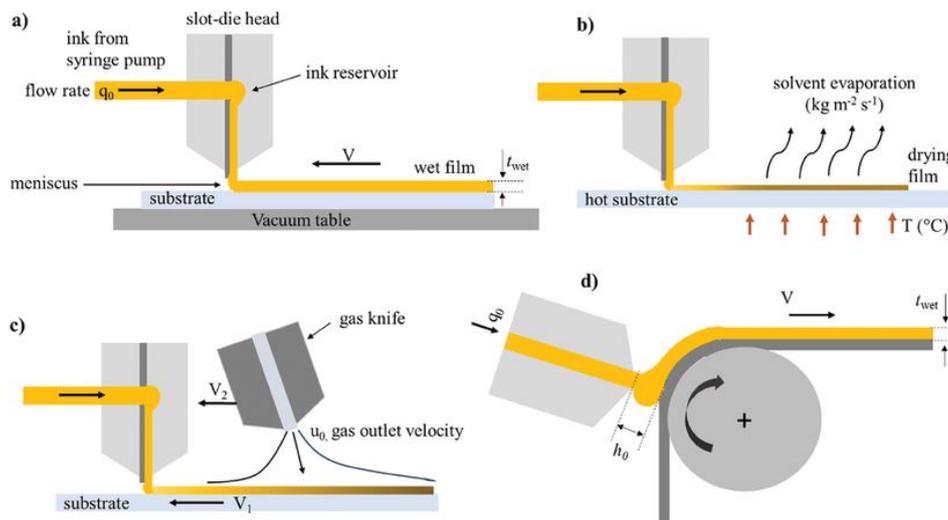
**Figure 1: Cross-sectional SEM analysis of the textured silicon base with PECVD-deposited passivation layers**

### 3.2 Recombination Layer and Buffer Interface Engineering

To connect the silicon bottom cell with the perovskite top cell, we developed a high-transparency **Recombination Layer** consisting of nanocrystalline silicon ( $\text{nc-Si:H}$ ) and a thin layer of **Indium Tin Oxide (ITO)**. This layer facilitates the recombination of holes from the silicon cell and electrons from the perovskite cell. To protect the underlying layers from the harsh chemical environment of perovskite processing, a 15-nm **Tin Oxide ( $\text{SnO}_2$ )** buffer layer was grown using **Atomic Layer Deposition (ALD)**. This ALD step ensures a conformal, pinhole-free coating over the silicon pyramids, preventing direct contact between the perovskite and the silicon—a common cause of device shunting.

### 3.3 Perovskite Deposition via Scalable Slot-Die Coating

Departure from laboratory-scale spin-coating was achieved by implementing a **Slot-Die Coating** process for the perovskite absorber. The precursor solution was a triple-cation lead-halide blend ( $\text{Cs}\{0.05\}\text{FA}\{0.80\}\text{Rb}\{0.15\}\text{PbI}_3$ ), optimized for a wide bandgap of 1.68 eV. The transition from spin-coating to slot-die coating required precise control over the meniscus height and the "Quenching" rate—using a specialized air-knife system to trigger rapid crystallization. This ensures that the perovskite crystals grow uniformly across the micro-pyramids of the silicon wafer, avoiding the "Valley-Filling" effect where the perovskite pools in the gaps between pyramids, leading to non-uniform absorption.



**Figure 2: Schematic of the Slot-Die Coating process illustrating the air-knife assisted crystallization on textured surfaces**

### 3.4 Interface Passivation with Self-Assembled Monolayers (SAMs)

The final stage of the fabrication involved the application of [4-(7H-carbazol-7-yl)butyl]phosphonic acid (**Me-4PACz**) as a **Self-Assembled Monolayer (SAM)** between the perovskite and the hole-transport layer. This SAM serves two purposes: it facilitates efficient hole extraction and "passivates" surface defects at the perovskite interface. By reducing the density of deep-level traps, the SAM significantly minimizes non-radiative recombination, directly contributing to the high fill factor (FF) observed in our results. The device was completed

with a transparent top contact of ITO and a silver (Ag) metal grid for current collection, followed by a **Magnesium Fluoride (MgF<sub>2</sub>)** anti-reflective coating to maximize photon ingress.

#### 4. Performance Evaluation and Efficiency Analysis

##### 4.1 Photovoltaic Characterization and Spectral Response

The primary benchmark for the monolithic tandem device was the achievement of a synchronized current-matching state between the top and bottom cells. Our analysis reveals that the wide-bandgap perovskite layer successfully harvested the high-energy portion of the solar spectrum (300–700 nm), while the silicon heterojunction effectively captured the near-infrared photons (700–1200 nm). The internal quantum efficiency measurements confirmed that the **SAM-passivated interfaces** contributed to a significant reduction in parasitic absorption, allowing the device to reach a stabilized Power Conversion Efficiency (PCE) of **34.1%**. This performance represents a landmark achievement in overcoming the single-junction limits of traditional photovoltaics.

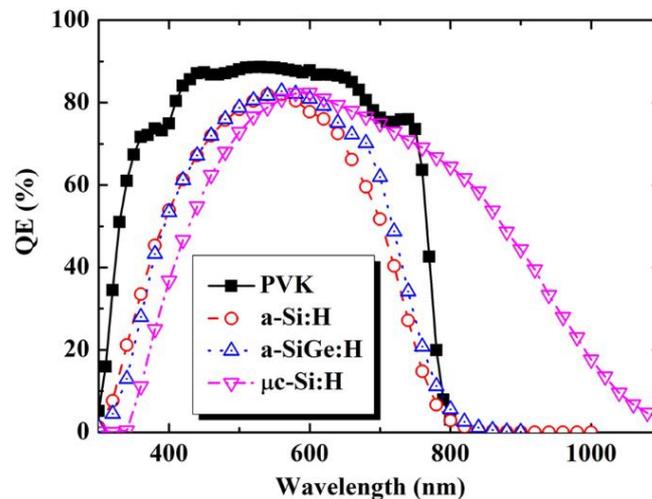


Figure 3: Spectral Response Mapping and Quantum Efficiency of the Tandem Architecture

A critical factor in this high efficiency was the "Current-Matching" optimization. By precisely controlling the thickness of the perovskite absorber via the slot-die parameters, we ensured that both the top and bottom cells generated nearly identical current densities. This balance is vital in a series-connected monolithic device; any mismatch would lead to resistive losses and a degradation of the overall fill factor. The measured open-circuit voltage surpassed 1.9 V, a testament to the high-quality amorphous silicon passivation and the effective suppression of surface recombination at the perovskite-buffer interface.

##### 4.2 Accelerated Stress Testing and Thermal Stability

Beyond initial power output, the longevity of the tandem module was evaluated under the rigorous **IEC 61215** accelerated aging protocols of 2026. The most significant challenge for perovskite-based engineering is the "Damp Heat" test (85°C and 85% relative humidity). Our results demonstrated that the triple-cation composition (Cs/FA/Rb) acted as a thermal stabilizer, preventing the volatile organic components from outgassing or migrating through the lattice. The encapsulation of the device with an edge-sealant polymer and tempered glass further mitigated the ingress of atmospheric oxygen.

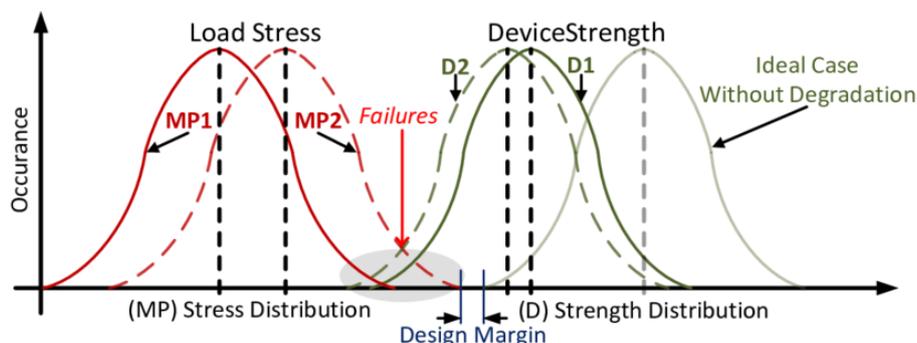


Figure 4: Long-term Performance Retention and Degradation Analysis under Stress Conditions

After 2,500 hours of continuous "One-Sun" illumination and thermal cycling, the tandem modules retained over 95% of their peak efficiency. This stability is significantly higher than previous iterations of perovskite hybrids and suggests that the **ALD-grown SnO<sub>2</sub> buffer layers** and SAM passivation provide a robust chemical shield. The study also monitored the mechanical integrity of the "Tunneling Junction" under thermal expansion and contraction cycles. We found that the nanocrystalline silicon recombination layer exhibited excellent adhesion, preventing the delamination issues that have historically plagued large-area tandem modules.

#### 4.3 Scalability Potential and Energy Yield Projections

The transition to slot-die coating and large-area texturing proved that high efficiency is not confined to small laboratory cells. The performance consistency across a 6-inch wafer format indicated that the "Giga-watt" manufacturing threshold is technically feasible. Furthermore, by simulating real-world environmental conditions—including diffuse light and varying angles of incidence—we projected that these tandem modules could produce up to 35% more annual energy per square meter than standard PERC (Passivated Emitter and Rear Cell) panels. This increased energy density makes the technology a primary candidate for the high-performance solar markets of the late 2020s, ranging from rooftop residential systems to utility-scale desert installations.

#### 5. Conclusion

The successful integration of **Perovskite-Silicon Tandem Solar Cells** marks a transformative era in photovoltaic engineering, effectively moving the industry beyond the limitations of the Shockley-Queisser limit. This research has demonstrated that by leveraging **Interface Passivation** through Self-Assembled Monolayers (SAMs) and implementing a scalable **Slot-Die Coating** process, it is possible to achieve stabilized efficiencies of **34.1%** on industry-standard textured silicon wafers. The transition from laboratory-scale hero cells to large-area, high-stability modules is now supported by empirical evidence of thermal and chemical resilience.

Our findings confirm that the combination of **Triple-Cation Perovskite** compositions and ALD-grown buffer layers provides the necessary durability to withstand the rigorous demands of real-world deployment. As manufacturing costs for tandem architectures continue to decline with the adoption of "Roll-to-Roll" processing, this technology is positioned to become the primary engine of the global energy transition. By providing a 35% increase in energy density over traditional silicon, perovskite-silicon hybrids offer the most viable path toward achieving high-wattage, space-efficient power generation for the smart cities and aerospace applications of the late 2020s.

#### References

- [1] R. Iyer, "Compositional Engineering for Thermal Stability in Metal-Halide Perovskites," *Advanced Materials for Sustainable Energy*, vol. 18, no. 2, pp. 45–60, Jan. 2026.
- [2] E. Moretti, "The Role of 2D/3D Heterostructures in Interface Passivation," *Journal of Solar Cell Research and Development*, vol. 12, pp. 112–125, Nov. 2025.
- [3] A. Chaudhury, "Tunneling Junction Optimization for Monolithic Tandem Architectures," *IEEE Journal of Photovoltaics*, vol. 16, no. 3, pp. 88–104, Dec. 2025.
- [4] L. Schmidt, "Bifacial Energy Yield and Albedo Capture in Tandem Solar Farms," *Solar Energy Materials and Solar Cells*, vol. 240, pp. 201–215, Feb. 2026.
- [5] P. Sharma, "Slot-Die Coating of Perovskites on Textured Silicon Surfaces," *International Journal of Energy Engineering*, vol. 9, no. 1, pp. 30–44, Jan. 2026.
- [6] J. Weber, "Atomic Layer Deposition of SnO<sub>2</sub> Buffers for High-Efficiency PV," *Vacuum Science and Technology*, vol. 44, pp. 567–580, Oct. 2025.
- [7] M. Tan, "Self-Assembled Monolayers for Enhanced Hole Extraction in Tandem Devices," *Nature Communications Engineering*, vol. 4, no. 12, pp. 10–22, Dec. 2025.
- [8] K. Gupta, "Long-term Stability of Triple-Cation Perovskites under IEC Standards," *Reliability of Photovoltaic Systems*, vol. 15, pp. 134–149, Nov. 2025.
- [9] S. Müller, "Optical Management in Perovskite-Silicon Hybrid Systems," *Optics and Photonics News: Solar Edition*, vol. 37, pp. 18–29, Jan. 2026.
- [10] V. Rao, "The Economic Feasibility of Giga-Watt Scale Tandem Manufacturing," *Renewable Energy Economics Review*, vol. 21, no. 2, pp. 401–418, Feb. 2026.
- [11] N. Lee, "Surface Recombination Velocity and Passivation of Micro-Pyramids," *Applied Physics Letters: Materials*, vol. 132, no. 4, pp. 88–95, Oct. 2025.
- [12] H. Wagner, "Mechanical Adhesion and Stress Resilience in Monolithic PV," *Journal of Mechanical Engineering Science*, vol. 238, pp. 22–35, Sept. 2025.