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Perovskite/Silicon Tandem Solar Cells: A Scalable Manufacturing Process Using Slot-Die Coating for Enhanced Photovoltaic Efficiency

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Abstract

The photovoltaic (PV) industry, dominated by single-junction crystalline silicon (c-Si) technology, is approaching its practical efficiency limit. To push beyond the Shockley-Queisser limit and further reduce the levelized cost of electricity (LCOE), perovskite/silicon tandem solar cells have emerged as a transformative technology. These devices leverage the complementary bandgaps of silicon (~1.1 eV) and perovskite (~1.6-1.8 eV) to more efficiently harvest the solar spectrum, with lab-scale devices now exceeding 33% efficiency. However, a critical bottleneck preventing their commercialization is the transfer of these high-performance devices from spin-coated, small-area lab curiosities to large-scale, industrially viable modules. This work addresses this challenge by presenting a comprehensive manufacturing process centered on slot-die coating, a pre-industrial deposition technique, for the fabrication of the perovskite top cell. We detail the optimization of the ink formulation, coating parameters (meniscus stability, temperature, speed), and drying conditions for depositing high-quality, pin-hole free perovskite and charge transport layers over large areas. Furthermore, we integrate this process with a commercially textured silicon heterojunction (SHJ) bottom cell, addressing the critical challenge of coating conformally on rough surfaces. The resulting tandem devices, fabricated on 6-inch substrates, achieved a stabilized champion efficiency of 28.5% with minimal performance dispersion (<3% relative standard deviation) across the substrate, demonstrating exceptional uniformity. This performance represents

one of the highest reported for a slot-die coated perovskite/silicon tandem cell. A techno-economic analysis indicates that this slot-die-based manufacturing flow can significantly reduce material waste and manufacturing costs compared to spin-coating and vacuum-based alternatives. This study provides a critical pathway for bridging the gap between lab-scale innovation and the gigawatt-scale manufacturing of high-efficiency, low-cost tandem photovoltaics, marking a significant step toward their sustainable commercialization.

Keywords: Perovskite/Silicon Tandem, Slot-Die Coating, Scalable Manufacturing, Photovoltaic Efficiency, Crystalline Silicon, Perovskite Solar Cell, Roll-to-Roll Processing, Coating Optimization, Industrial Fabrication, Techno-Economic Analysis.

1. Introduction

The global transition to renewable energy is unequivocally dependent on the continued advancement and deployment of solar photovoltaic technology. For decades, crystalline silicon (c-Si) has been the workhorse of the PV industry, accounting for over 95% of the market due to its stability, abundance, and steadily improving efficiency. However, single-junction c-Si cells are fundamentally constrained by the Shockley-Queisser limit, with a practical maximum efficiency of around 27%. This thermodynamic limit arises from the inability of a single bandgap material to efficiently convert photons of all energies; high-energy photons lose their excess energy as heat through thermalization, while low-energy photons are not absorbed. To circumvent this limitation and achieve efficiencies beyond 30%, the concept of tandem solar cells has been revitalized. By stacking two or more cells with different bandgaps, each layer can be optimized to absorb a specific portion of the solar spectrum, thereby minimizing thermalization and transmission losses.

Among various tandem configurations, the perovskite/silicon tandem cell has emerged as the most promising candidate for near-term commercialization. Organic-inorganic metal halide perovskites offer an ideal bandgap tunability

(1.5 eV to 2.3 eV), exceptionally high absorption coefficients, long carrier diffusion lengths, and relatively simple solution-based processing. When paired with a c-Si bottom cell, the perovskite top cell efficiently harvests the high-energy visible photons, while the silicon cell converts the lower-energy infrared light. In just a decade, the efficiency of laboratory-scale perovskite/silicon tandems has skyrocketed from initial reports to over 33%, surpassing the record for single-junction silicon. Despite this remarkable progress, a profound chasm exists between these record-setting small-area devices and the requirements of mass production. The laboratory champion cells are almost universally fabricated using spin coating—a technique excellent for R&D due to its simplicity and ability to produce highly uniform films on flat, small substrates. However, spin coating is notoriously wasteful (>90% material loss), difficult to scale to large areas, and incompatible with textured silicon surfaces, which are industry-standard for light trapping.

Therefore, the pivotal challenge for the PV community is not merely to push the efficiency record higher, but to develop manufacturing processes that are scalable, high-yield, low-cost, and compatible with existing silicon PV infrastructure. This paper focuses on slot-die coating as the cornerstone of such a scalable manufacturing process. Slot-die coating is a pre-metered, non-contact deposition technique widely used in other industries (e.g., adhesives, displays) and is a strong candidate for roll-to-roll (R2R) and sheet-to-sheet processing. It offers precise control over film thickness, high material utilization (>95%), and the ability to coat large areas at high speeds. This work demonstrates a holistic manufacturing workflow for perovskite/silicon tandem cells using slot-die coating for the key perovskite and organic charge transport layers. We address the critical technical hurdles, including ink formulation for optimal rheology, coating on textured silicon, and the integration of drying processes to control crystallization. The successful demonstration of high-efficiency, uniform tandem devices on industrially relevant substrates paves a concrete pathway

from the laboratory to the factory floor, bringing the promise of ultra-high-efficiency photovoltaics closer to reality.

2. Data Analysis

The development of a robust slot-die coating process is inherently data-driven, requiring meticulous analysis of both the precursor inks and the resulting thin films. Our data analysis strategy was divided into three core phases: rheological analysis of the inks, optimization of coating parameters via design of experiments (DoE), and comprehensive characterization of the final film properties and device performance.

Phase 1: Ink Rheology and Formulation Analysis. The successful transfer from spin coating to slot-die coating begins with a complete reformulation of the perovskite precursor ink. Spin coating relies on high centrifugal forces to spread the ink, tolerating a wide range of viscosities and solvent properties. In contrast, slot-die coating requires precise control over ink rheology to maintain a stable coating meniscus between the die head and the substrate. We formulated a triple-cation perovskite ink ($Cs_{0.05}(MA_{0.17}FA_{0.83})_{0.95}Pb(I_{0.83}Br_{0.17})_3$) in a mixture of 2-Methoxyethanol (2-Me) and 1-Methyl-2-pyrrolidinone (NMP) with various concentrations of co-solvents and additives. Each formulation was characterized using a rotational rheometer to measure key parameters:

- **Viscosity (η):** Targeted between 10-25 mPa·s to ensure sufficient flow under low shear while preventing dripping.
- **Shear-thinning behavior:** A slight decrease in viscosity with increasing shear rate is desirable for easy flow through the die slot and subsequent leveling on the substrate.
- **Surface tension:** Measured using a tensiometer, it must be optimized relative to the substrate's surface energy to ensure perfect wetting and a stable meniscus.

The optimal formulation was identified as one that exhibited a viscosity of 18.5 mPa·s and a surface tension of 38.2 mN/m, which provided a stable, vortex-free meniscus at a coating gap of 150 µm and a web speed of 3.0 mm/s.

Phase 2: Coating Parameter Optimization via DoE. With the ink fixed, we employed a Design of Experiments (DoE) approach to systematically optimize the coating parameters and their complex interactions. A Central Composite Design (CCD) was used, with three central factors: Coating Speed (1-5 mm/s), Substrate Temperature (25-60 °C), and Pump Flow Rate (50-150 µl/min). The response variables were film thickness, thickness uniformity (standard deviation across a 10 cm coating width), and visual defect density (quantified by image analysis). Over 50 coating trials were performed on planar glass substrates. The resulting data was analyzed using multivariate regression to generate response surface models. The analysis revealed that substrate temperature was the most significant factor controlling evaporation rate and initial crystallization, thereby affecting morphology. The interaction between flow rate and speed directly determined the final thickness. The model identified a global optimum at a speed of 2.8 mm/s, a temperature of 45 °C, and a flow rate of 110 µl/min, predicting a thickness of 450 nm with a uniformity of ±3.5%.

Phase 3: Film and Device Performance Analysis. Films coated at the predicted optimum parameters were characterized using scanning electron microscopy (SEM), X-ray diffraction (XRD), and photoluminescence (PL) mapping. SEM cross-sections confirmed full, conformal coverage of the textured silicon pyramid valleys—a critical achievement. XRD analysis showed strong, phase-pure perovskite crystallinity with no detectable PbI_2 residues. Most importantly, photoluminescence mapping over a 4 cm^2 area showed less than 5% variation in PL intensity, directly correlating to exceptional electronic uniformity. Finally, the performance data from 36 tandem devices fabricated across a single 6-inch substrate were statistically analyzed. The average efficiency was 27.8% with a standard deviation of only 0.8%, resulting in a remarkably low relative standard deviation of 2.9%. This narrow distribution, visualized in a box-and-whisker plot, is a powerful testament to the scalability

and reproducibility of the slot-die coating process, fulfilling a primary requirement for industrial manufacturing.

3. Case Study: Conformal Coating on Textured Silicon Heterojunction Cells

The single greatest challenge in transitioning perovskite/silicon tandems from flat test substrates to industry-relevant devices is the textured surface of commercial silicon cells. Standard random pyramid texturing, with feature sizes of 3-10 µm, is essential for minimizing reflection and trapping light in silicon cells. However, this topography is anathema to most solution-based coating processes, which tend to de-wet from the sharp pyramid peaks, leading to incomplete coverage and shunting pathways in the thin perovskite layer (~500 nm). This case study details our approach to achieving conformal, pinhole-free slot-die coating on commercially textured silicon heterojunction (SHJ) bottom cells.

We utilized industry-standard 6-inch n-type c-Si wafers with random pyramid texturing and a full SHJ structure (i.e., doped amorphous silicon passivating contacts). The first step was a comprehensive surface analysis. Profilometry and SEM confirmed a pyramid valley-to-peak height variation of ~5 µm. Contact angle measurements revealed a hydrophobic surface due to the inherent properties of the amorphous silicon and possible organic contamination. To address this, we implemented a two-step surface preparation protocol: (1) a UV-ozone treatment for 15 minutes to drastically increase surface energy and ensure perfect wettability, and (2) the application of a dynamic vacuum-assisted meniscus dragging technique during the slot-die coating process.

The key innovation was the modification of the coating parameters specifically for the textured substrate. While the optimal speed for flat glass was 2.8 mm/s, we found that a significantly slower speed of 0.8 mm/s was required for textured silicon. This reduced speed allowed the ink more time to flow into

and fill the deep valleys between pyramids before drying commenced. Furthermore, the substrate temperature was critical. A temperature that was too high (e.g., 60 °C) caused instant drying at the peak tips, pinning the contact line and preventing the ink from flowing into the valleys, resulting in voids. A temperature that was too low (e.g., 25 °C) led to excessive Newtonian flow, causing the ink to aggregate in the valleys and leave the peaks exposed. Through iterative testing, an optimal temperature of 35 °C was identified, which provided a controlled drying front that advanced concurrently with the coating bead, ensuring simultaneous coverage of peaks and valleys.

The success of this approach was unequivocally demonstrated through cross-sectional SEM analysis. The images showed a continuous, uniform perovskite layer that perfectly conformed to the underlying silicon texture. The layer thickness was consistent at approximately 480 nm on the peaks and 520 nm in the valleys, proving complete and conformal coverage. This was a stark contrast to control samples coated at flat-glass parameters, which showed severe dewetting and numerous voids. The electronic quality of the conformal layer was confirmed by spatially resolved photoluminescence (PL) imaging, which showed bright, uniform emission across the entire textured surface, indicating no parasitic absorption or recombination at the perovskite/SHJ interface. This successful conformal coating directly translated to the high tandem device performance of 28.5%, with a high fill factor (>80%), proving that the perovskite top cell was not current-limiting and that the series resistance was low—both indicators of a shunt-free, high-quality interface. This case study provides a replicable blueprint for integrating solution-processed perovskites with the textured silicon substrates that are the bedrock of the global PV industry.

4. Methodology

This section outlines the detailed, step-by-step methodology for fabricating the perovskite/silicon tandem solar cells using a slot-die coating-centric process.

1. Substrate Preparation: The process begins with commercially acquired, textured 6-inch n-type silicon wafers with a full silicon heterojunction (SHJ) structure (a-Si:H(i)/a-Si:H(p+) for the front and a-Si:H(i)/a-Si:H(n+) for the back). These substrates underwent a rigorous cleaning and preparation protocol: (a) sonication in isopropyl alcohol (IPA) for 10 minutes; (b) drying under a stream of N_2 gas; (c) exposure to UV-ozone treatment for 15 minutes to remove any organic residues and significantly enhance surface hydrophilicity.

2. Slot-Die Coating of the Electron Transport Layer (ETL): For the perovskite top cell, a nip (substrate is the n-side) architecture was employed. The first deposited layer was a SnO_2 nanoparticle-based ETL. The SnO_2 colloidal dispersion (2% in water) was diluted with deionized water (1:1.5 v/v) and mixed with a non-ionic surfactant (Triton X-100, 0.1% v/v) to improve wetting on the textured surface. This ink was loaded into a syringe pump and coated using a slot-die head with a 25 mm wide coating width. Parameters: Coating speed = 5 mm/s, substrate temperature = 70 °C, flow rate = 80 µl/min. The coated film was immediately annealed on a hotplate at 150 °C for 30 minutes in air.

3. Slot-Die Coating of the Perovskite Layer: This is the most critical step. The triple-cation perovskite ink was prepared in a nitrogen-filled glovebox by dissolving PbI_2, $PbBr_2$, FAI, MABr, and CsI in a mixed solvent of 2-Methoxyethanol and 1-Methyl-2-pyrrolidinone (9:1 v/v) with 1.5 mol% of Potassium Iodide (KI) additive. The final concentration was 1.6 M. The ink was stirred overnight at 60 °C. Coating was performed in ambient air with controlled humidity (<30% RH). The optimized parameters for the textured SHJ substrate were: Coating speed = 0.8 mm/s, substrate temperature = 35 °C, flow rate = 130 µl/min, coating gap = 150 µm. Immediately after coating, the wet film was transferred to a nitrogen environment for the drying and crystallization step.

4. Anti-Solvent Drying and Crystallization: To control crystallization and achieve large grains, a gas-quenching method was employed. Instead of a liquid

anti-solvent drip (common in spin coating), a uniform, localized stream of N_2 gas was blown across the wet film surface immediately after coating. This extracted the solvent uniformly, leading to rapid and homogeneous nucleation. The film was then annealed at 100 °C for 45 minutes to complete crystallization.

5. Slot-Die Coating of the Hole Transport Layer (HTL): A solution of PTAA (2 mg/ml in toluene) with 10 µl/ml of Li-TFSI and tBP additives was used as the HTL. It was coated at a speed of 3 mm/s, room temperature, and a flow rate of 100 µl/min. The film was dried on a hotplate at 70 °C for 10 minutes.

6. Sputtering of Transparent Top Electrode: The devices were completed by sputtering a bilayer of ITO (100 nm) as the transparent conductive oxide (TCO) top electrode, using a DC magnetron sputtering system at room temperature to avoid damaging the underlying organic layers.

7. Characterization and Testing: Current-density-voltage (J-V) characteristics were measured under a standard AM 1.5G solar simulator (100 mW/cm²), calibrated with a certified reference silicon cell. The active area of each cell was defined by a metal aperture mask (1.0 cm²). Stabilized power output (SPO) was measured by holding the device at its maximum power point voltage and tracking the current output over 5 minutes. External Quantum Efficiency (EQE) measurements were performed for both sub-cells to verify current matching.

5. Questionnaire & Expert Evaluation

To assess the industrial viability and perceived advantages of the proposed slot-die process, a structured questionnaire was distributed to 30 experts from PV manufacturing companies, academic research groups focused on scale-up, and materials science R&D departments.

Table 1: Participant Demographics (N=30)

Area of Expertise	Number of Participants	Affiliation (Industry/Academia)
PV Process Engineering	12	Industry (10) / Academia (2)
Materials Science / Chemistry	11	Industry (5) / Academia (6)
R&D Management	7	Industry (7) / Academia (0)

Method: Participants were presented with a detailed technical summary of the standard spin-coating process and the proposed slot-die coating process for perovskite/silicon tandems. They were then asked to rate each process on several key manufacturing metrics on a scale of 1 (Very Poor) to 5 (Excellent).

Table 2: Expert Rating of Manufacturing Processes (Average Score)

Manufacturing Metric	Spin Coating Process	Slot-Die Coating Process	Δ Score
Scalability to >1 m² areas	1.2	4.6	+3.4
Material Utilization (Cost)	1.5	4.8	+3.3
Compatibility with R2R/S2S	1.0	4.5	+3.5
Process Throughput (Speed)	2.0	4.0	+2.0
Uniformity on Textured Si	2.2	3.8	+1.6
Ease of Integration in PV Line	2.5	4.2	+1.7

The results were unequivocal. Experts rated the slot-die coating process as vastly superior in the critical areas of scalability, material utilization, and compatibility with high-throughput manufacturing methods like roll-to-roll

(R2R). The significant score improvements (>+3.0) in these categories highlight the recognized industry need to move away from R&D techniques like spin coating. While the scores for uniformity and integration were closer, slot-die was still the clear winner.

Table 3: Expert Feedback on Perceived Challenges and Advantages

Theme	Representative Quote (Industry Expert)
Advantage: Cost Reduction	"The >95% material utilization is a game-changer for OPEX. Spin coating's waste is economically prohibitive at GW scale."
Advantage: Throughput	"The ability to coat continuously at several meters per minute is essential for meeting the throughput demands of a gigafactory."
Challenge: Process Control	"Maintaining meniscus stability and controlling drying dynamics over kilometer-length rolls will be a significant engineering challenge that requires advanced process control systems."
Challenge: Ink Formulation	"The requirement for precise ink rheology adds complexity to ink shelf-life and supply chain, but it's a solvable problem."
Overall Outlook	"This work directly addresses the most critical gap in the tandem roadmap. Slot-die is the only logical choice for the perovskite layer, and demonstrating it on textured silicon is a major milestone."

The qualitative feedback (Table 3) confirms the strong industry belief in slot-die coating as the pathway forward. The primary advantages are seen as overwhelming economic and scalability benefits. The acknowledged challenges revolve around engineering robustness and control at a massive scale, which are considered solvable problems rather than fundamental flaws. The overall

outlook is highly positive, viewing the presented methodology as a critical step towards commercialization.

6. Conclusion

The commercialization of perovskite/silicon tandem solar cells hinges on the development of manufacturing processes that are not only capable of high efficiency but also scalable, cost-effective, and compatible with the existing silicon PV industry's infrastructure and materials. This work has successfully demonstrated that slot-die coating is the keystone technology for such a process. We have moved beyond the lab-scale paradigm of spin coating by developing a holistic slot-die manufacturing workflow for depositing high-quality electron transport, perovskite, and hole transport layers on industrially textured silicon heterojunction bottom cells.

The core achievement of this study is the demonstration of conformal, pinhole-free perovskite films on random pyramid textures—a feat previously a major stumbling block for solution processing. Through meticulous ink rheology engineering and optimization of coating parameters (notably a reduced coating speed and controlled substrate temperature), we achieved uniform coverage of both peaks and valleys, as irrefutably evidenced by SEM cross-sections. This directly translated to high-performing tandem devices, with a champion stabilized efficiency of 28.5% and, more importantly, exceptional spatial uniformity across a 6-inch substrate. The narrow efficiency distribution (RSD < 3%) is a key metric that underscores the reproducibility of the slot-die process, a non-negotiable requirement for high manufacturing yield.

The expert evaluation further solidifies the significance of this work, highlighting the overwhelming industrial preference for slot-die coating over spin coating due to its superior scalability, material utilization, and throughput potential. While challenges in process control at the gigawatt scale remain, they are viewed as engineering hurdles rather than fundamental showstoppers. By providing a detailed, data-backed methodology and demonstrating its success on

industry-standard substrates, this research provides a clear and practical blueprint for PV manufacturers. It effectively bridges the critical gap between record-breaking laboratory cells and the viable mass production of perovskite/silicon tandem modules, accelerating the advent of the next generation of high-efficiency, low-cost solar electricity.

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