

## Supporting Information

### **Scalable deposition and drying methods toward large-area monolithic perovskite/silicon tandem solar cells**

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**Table S1** Summary of scalable deposition methods for perovskite films in monolithic PSTSCs: working principle, silicon compatibility, film quality, reliability, and industrial potential

Deposition Method	Working Principle	Silicon Compatibility (textured Si bottom cells)	Process Complexity	Film Quality	Implementation Reliability	Cost Analysis( <i>I</i> )	Industrial Potential	Advantages	Limitations / Challenges
Blade Coating	Solution-based, spreading precursor solution with a blade over substrate	Moderate; wetting on textures may require surface modification or texture modulation	Low	Film thickness control achievable with optimization	High for small to medium areas; challenging for very large areas	CAPEX: 0.30-0.33 \$/W, low OPEX	High: compatible with R2R and large-area deposition	Simple, scalable, low-cost	Thickness uniformity over very large areas can be challenging; prone to defects if solvent evaporation not controlled
D-bar Coating	Solution-based, similar to blade coating but uses a bar	Moderate; similar wetting issues as blade coating	Low	Similar as blade coating	Moderate	CAPEX: 0.30-0.33 \$/W, low OPEX	High	Simple, fast coating; compatible with large-area substrates	Sensitive to solution rheology and substrate wetting
Slot-Die Coating	Solution-based, precise metering through a die slot for continuous coating	Moderate; adapt to textured surfaces with careful flow control	Moderate	Good thickness uniformity for large-area films	High; stable process with proper flow control	CAPEX: 0.30-0.33 \$/W, low OPEX	Very high; compatible with R2R manufacturing	High throughput, scalable, reproducible	Requires precise flow rate and substrate alignment; complex setup
Spray Coating	Solution-based, atomized droplets deposited on substrate	Good; non-uniform deposition on deep textures possible	Moderate	Depends on droplet size and substrate interaction	Moderate; droplet consistency is critical	CAPEX: 0.30-0.33 \$/W, low to moderate OPEX	Moderate; scalable but may need multiple passes	Flexibility in substrate shape; scalable	Droplet coalescence can cause defects; control over thickness challenging
Inkjet Printing	Solution-based, digital droplet deposition	Good; wetting issues on textured surfaces	Moderate to high	Good for small areas; may struggle with large-area uniformity	Moderate; prone to clogging and nozzle variations	CAPEX: slightly higher than other solution methods, low OPEX	Moderate; suitable for patterned deposition, less for full-area coating	Precision patterning, material saving	Limited throughput; nozzle reliability; difficult for very large areas
Screen Printing	Solution/paste-based, stencil-assisted deposition	Moderate; mechanical contact may damage delicate textures	Moderate	Film uniformity depends on mesh and paste	High; robust for thick layers	CAPEX: 0.30-0.33 \$/W, low OPEX	High; widely used for large-area electrodes	Simple, robust, scalable	Limited to thicker films; resolution limited

<b>Deposition Method</b>	<b>Working Principle</b>	<b>Silicon Compatibility (textured Si bottom cells)</b>	<b>Process Complexity</b>	<b>Film Quality</b>	<b>Implementation Reliability</b>	<b>Cost Analysis(1)</b>	<b>Industrial Potential</b>	<b>Advantages</b>	<b>Limitations / Challenges</b>
Evaporation	Simultaneous or sequential vapor-phase deposition of multiple precursors	High; compatible with textured substrates	High	Dense, uniform films but requires complex optimization	High; requires precise rate control	CAPEX: ~1 \$/W, moderate OPEX	Moderate; mostly batch-based	Precise stoichiometry, uniform films	Very high capital cost; complex process integration
Hybrid deposition	Sequential: inorganic framework deposited by vapor, followed by solution-based organic salts to form perovskite	High; inorganic template compatible with textures	Moderate	Dense, uniform crystalline films achievable, but the organic salt requires suitable proportion	Moderate; reproducibility sensitive to solution and environment	CAPEX: ~0.3-1 \$/W, moderate OPEX for solution	Moderate; scalable with process integration	Combines precision of inorganic control with solution-based crystallization	Complex sequential process; sensitive to environment

CAPEX: Capital Expenditure; OPEX: Operating Expenditure

**Table S2** Representative metrics currently reported in the literature for evaluating the industrial reliability of manufacturing perovskite layers in monolithic PSTSCs using different scalable deposition techniques

<b>Deposition Method</b>	<b>Reported area (cm<sup>2</sup>)</b>	<b>Reported Lifespan (h)</b>	<b>Compatibility with M12-size substrates</b>	<b>References</b>
Blade Coating	24	T <sub>80</sub> =1700	yes	(2-4)
D-bar Coating	-	-	yes	(5)
Slot-die Coating	260	T <sub>87.7</sub> =1500	yes	(6)
Spray Coating	-	-	yes	(7)
Inkjet Printing	-	-	yes	(8)
Screen Printing	-	-	yes	(9)
Vapor-phase deposition	1	T <sub>90</sub> =2164	yes	(10)
Hybrid deposition	64.64	T <sub>100</sub> =1000	yes	(11, 12)
Unknown	330.56/274.1	-	-	(13)

M12-size: 210×210 mm<sup>2</sup>

Due to the lack of standardized industrial metrics such as throughput and yield, the evaluation is currently based on representative areas, lifespans, and compatibility with M12-size substrates.

**Table S3** Summary of scalable drying techniques for perovskite films in monolithic PSTSCs: working principles, kinetics control, and scalability considerations

Drying Method	Working Principle	Drying Dynamics / Kinetics Control	Film Quality	Implementation Reliability	Process Complexity	Cost Analysis	Industrial Potential	Key Advantages	Challenges and Limitations
Anti-solvent Extraction	Antisolvent displaces host solvent, triggering fast crystallization.	Extremely fast nucleation; rapid solvent exchange; difficult to scale due to droplet dynamics.	High-quality, large-grained films	Low; limited large-area applications	Moderate-to-high; requires solvent management and waste recovery.	Low CAPEX (simple delivery systems), moderate (consumable chemicals, waste handling)	Limited to small-scale or spin-based processes.	Produces smooth, high-performance films in small areas.	Poor scalability; solvent toxicity and waste issues.
Substrate Preheating	Substrate preheated to accelerate solvent evaporation and induce intermediate-phase formation.	Moderate drying rate; temperature-dependent nucleation control.	Uniformity control for large area	Highly reproducible and easy to control; minimal environmental dependence.	Low; requires only temperature control.	Low CAPEX (heaters/IR lamps), moderate (energy usage)	Easily integrated into existing coating tools.	Simple, low-cost, energy-efficient; enhances nucleation uniformity.	Risk of non-uniform heating on large substrates; not suitable for temperature-sensitive layers.
Gas Flow	A stream of inert gas (e.g., N <sub>2</sub> , Ar) accelerates solvent evaporation and controls nucleation.	Moderate to fast solvent removal; drying rate tunable by gas velocity and temperature; enables gradient-free crystallization.	Uniform, dense, pinhole-free films	High reproducibility under controlled flow; tolerant to ambient variations.	Moderate; requires fine tuning of gas direction, flow rate, and distance.	Moderate CAPEX (gas handling & nozzles), moderate (gas supply, utilities)	Highly scalable; easily integrated into continuous coating lines.	Non-contact, fast, and compatible with large-area manufacturing.	Sensitive to gas uniformity; excessive flow can cause surface roughening or premature crystallization.
Vacuum Quenching	Solvent rapidly evaporated under low pressure, inducing supersaturation and fast nucleation.	Fast solvent extraction; drives uniform intermediate-phase formation and dense crystal packing, but difficult to extract high boiling point solvents	Compact and smooth films with large grains and minimal voids.	Reliable under precise pressure and timing control; batch-to-batch stability achievable.	Moderate; requires vacuum chamber and timing match with coating step.	Moderate-High CAPEX (vacuum chamber), higher OPEX (vacuum pumps)	Scalable in inline or batch vacuum systems.	Produces dense films with strong crystallinity.	Equipment-intensive; limited continuous processing; higher energy load.
Radiative Heating (Infrared / Halogen)	IR or halogen radiation rapidly heats wet film to induce fast solvent removal.	Fast and controllable drying kinetics; heating intensity governs crystallization front propagation.	Dense, smooth films; improved optoelectronic quality.	High reproducibility with optimized lamp intensity and exposure time.	Moderate-to-high; requires uniform lamp distribution and precise thermal control.	Moderate CAPEX (heating arrays), moderate (energy/hours processed)	High potential for inline and continuous processing.	Rapid, contactless, and scalable; conformal drying.	Local overheating risk; lamp uniformity critical; needs real-time feedback control.

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