

# **SCALING UP PEROVSKITE SOLAR CELLS VIA MODULATING FUNCTIONAL LAYERS**

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**DEPARTMENT OF ENERGY SCIENCE AND ENGINEERING**

**INDIAN INSTITUTE OF TECHNOLOGY DELHI**

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*by*

**Nitin Kumar Bansal**

**DEPARTMENT OF ENERGY SCIENCE AND ENGINEERING**

*submitted*

*in fulfillment of the requirement of the degree of*

**Doctor of Philosophy**

*to the*



**INDIAN INSTITUTE OF TECHNOLOGY DELHI**

**JULY 2026**

*Dedicated*  
*To*  
*My Parents*

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## DECLARATION

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I certify that

- a. The work contained in the thesis is original and has been done by myself under the supervision of my supervisor.
- b. The work has not been submitted to any other Institute for any degree or diploma.
- c. I have followed the guidelines provided by the Institute in writing the thesis.
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Nitin Kumar Bansal

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## CERTIFICATE

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This is to certify that the thesis entitled “*Scaling Up Perovskite Solar Cells via Modulating Functional Layers*”, submitted by **Nitin Kumar Bansal** to the Indian Institute of Technology, Delhi, is a record of bona fide research work under my supervision, and I consider it worthy of consideration for the award of the degree of Doctor of Philosophy of the Institute. He has fulfilled all requirements for the submission of this thesis, which has attained the standard required for a Ph.D. degree of the Institute. The research reports and results presented in the thesis have not been submitted for any degree or diploma in any other university or institute.

Place:

Date:

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Nitin Kumar Bansal

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## ABSTRACT

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Organic–inorganic lead halide perovskite solar cells (PSCs) have emerged as a promising photovoltaic technology owing to their exceptional optoelectronic properties, including high absorption, long carrier diffusion lengths, and tunable bandgaps. Coupled with advances in composition engineering, additive incorporation, and interfacial modification, PSCs have achieved power conversion efficiencies (PCEs) exceeding 27%. Despite this rapid progress, most high-efficiency devices are fabricated under inert or tightly controlled environments, posing a significant challenge for scalable and cost-effective commercialization. The thesis addresses this gap by developing robust fabrication protocols aimed at scaling up PSCs under air-ambient conditions with controlled relative humidity (20–35%). Particular emphasis is placed on optimizing functional layers, including the electron transport layer (ETL) and the perovskite absorber, as well as their interfacial properties, which collectively govern device performance and operational stability. Compact TiO<sub>2</sub> and SnO<sub>2</sub> ETLs were systematically optimized and comprehensively characterized using various measurements, demonstrating that both materials can serve as effective ETLs for high-performance PSCs. Crystallization control of the perovskite layer was achieved through solvent and antisolvent engineering, enabling improved film morphology and reduced bulk defects. Further enhancement was realized through targeted interfacial engineering at the ETL/perovskite interface using the conjugated polyelectrolyte PFNBr, which promoted homogeneous grain growth, suppressed interfacial trap states, and facilitated more efficient electron extraction. As a result, optimized devices fabricated entirely in air exhibited PCEs exceeding 22% with high reproducibility (~90%). Complementary SCAPS-1D simulations were performed to elucidate the role of bilayer interfacial modification in defect passivation across a range of absorber properties, providing theoretical validation of the experimental observations. Additionally, functional layer optimization, combined with low-cost HTL-enabled devices, led to PCEs exceeding 23% and markedly improved operational stability. Finally, a scalable approach for large-area perovskite film fabrication (5 × 5 cm<sup>2</sup>) was established using a multichannel micropipette-assisted antisolvent dripping technique, yielding uniform, pinhole-free films over extended substrates, thereby advancing the prospects of PSCs toward FAB-scale.

**Keywords:** Perovskite solar cells, Interface engineering, Air ambient fabrication, Solvent engineering, Antisolvent engineering, Efficiency, Crystallization modulation, Stability

## सारांश (Abstract)

कार्बनिक-अकार्बनिक सीसा हैलाइड पेरोव्स्काइट सौर सेल (PSCs) अपनी उत्कृष्ट प्रकाश-वैद्युतिक (optoelectronic) विशेषताओं, जैसे उच्च अवशोषण क्षमता, लंबी वाहक प्रसार लंबाई (carrier diffusion length), तथा समायोज्य बैंडगैप के कारण एक अत्यंत आशाजनक फोटोवोल्टाइक तकनीक के रूप में उभरे हैं। संरचना अभियांत्रिकी, एडिटिव सम्मिलन तथा इंटरफेस संशोधन में प्रगति के साथ, PSCs ने 27% से अधिक शक्ति रूपांतरण दक्षता (PCE) प्राप्त कर ली है। हालांकि, इस तीव्र प्रगति के बावजूद, अधिकांश उच्च दक्षता वाले उपकरण निष्क्रिय (inert) या अत्यंत नियंत्रित वातावरण में निर्मित किए जाते हैं, जो उनके बड़े पैमाने पर एवं किफायती व्यावसायीकरण के लिए एक महत्वपूर्ण चुनौती प्रस्तुत करता है। यह शोध प्रबंध इस अंतर को दूर करने हेतु वायु परिवेश (air-ambient) में नियंत्रित सापेक्ष आर्द्रता (20–35%) के अंतर्गत PSCs के स्केलेबल निर्माण के लिए मजबूत निर्माण प्रक्रियाओं का विकास प्रस्तुत करता है। इस अध्ययन में कार्यात्मक परतों के अनुकूलन पर विशेष ध्यान दिया गया है, जिसमें इलेक्ट्रॉन परिवहन परत (ETL) और पेरोव्स्काइट अवशोषक परत, तथा इनके इंटरफेस गुण शामिल हैं, जो सामूहिक रूप से उपकरण के प्रदर्शन और संचालन स्थिरता को नियंत्रित करते हैं। कॉम्पैक्ट  $\text{TiO}_2$  तथा  $\text{SnO}_2$  ETLs का व्यवस्थित अनुकूलन और विभिन्न परीक्षणों द्वारा व्यापक विश्लेषण किया गया, जिससे यह प्रदर्शित हुआ कि दोनों सामग्री उच्च प्रदर्शन वाले PSCs के लिए प्रभावी ETL के रूप में कार्य कर सकती हैं। पेरोव्स्काइट परत की क्रिस्टलीकरण प्रक्रिया को सॉल्वेंट एवं एंटी-सॉल्वेंट अभियांत्रिकी के माध्यम से नियंत्रित किया गया, जिससे फिल्म की आकृति (morphology) में सुधार तथा थोक दोषों (bulk defects) में कमी प्राप्त हुई। इसके अतिरिक्त, संयुग्मित पॉलीइलेक्ट्रोलाइट PFNBr का उपयोग करते हुए ETL/पेरोव्स्काइट इंटरफेस पर लक्षित इंटरफेस अभियांत्रिकी द्वारा और अधिक सुधार प्राप्त किया गया, जिससे समान दाने (grain) वृद्धि को बढ़ावा मिला, इंटरफेस ट्रैप अवस्थाओं में कमी आई, तथा अधिक प्रभावी इलेक्ट्रॉन निष्कर्षण संभव हुआ। परिणामस्वरूप, पूर्णतः वायु परिवेश में निर्मित अनुकूलित उपकरणों ने 22% से अधिक PCE तथा उच्च पुनरुत्पादकता (~90%) प्रदर्शित की। इसके अतिरिक्त, SCAPS-1D सिमुलेशन के माध्यम से द्वि-परत इंटरफेस संशोधन की भूमिका का विश्लेषण किया गया, जिसने विभिन्न अवशोषक गुणों के संदर्भ में दोष निष्क्रियकरण (defect passivation) को स्पष्ट करते हुए प्रायोगिक परिणामों की सैद्धांतिक पुष्टि की। साथ ही, कार्यात्मक परतों के अनुकूलन तथा कम लागत वाले HTL आधारित उपकरणों के संयोजन से 23% से अधिक PCE तथा उल्लेखनीय रूप से बेहतर संचालन स्थिरता प्राप्त हुई। अंततः, मल्टीचैनल माइक्रोपिपेट-सहायित एंटी-सॉल्वेंट ड्रिपिंग तकनीक का उपयोग करते हुए बड़े क्षेत्र ( $5 \times 5$  सेमी<sup>2</sup>) की पेरोव्स्काइट फिल्म निर्माण की एक स्केलेबल विधि विकसित की गई, जिससे विस्तारित सबस्ट्रेट पर समान एवं पिनहोल-रहित फिल्म प्राप्त हुई, जो PSCs को FAB-स्तरीय निर्माण की दिशा में अग्रसर करती है।

**कुंजी शब्द (Keywords):** पेरोव्स्काइट सौर सेल, इंटरफेस अभियांत्रिकी, वायु परिवेश निर्माण, सॉल्वेंट अभियांत्रिकी, एंटी-सॉल्वेंट अभियांत्रिकी, दक्षता, क्रिस्टलीकरण नियंत्रण, स्थिरता

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## LIST OF SYMBOLS AND ABBREVIATIONS

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|                 |                                                                                                               |
|-----------------|---------------------------------------------------------------------------------------------------------------|
| PSC             | : Perovskite solar cell                                                                                       |
| ETL             | : Electron transport layer                                                                                    |
| HTL             | : Hole transport layer                                                                                        |
| TCO             | : Transparent conducting oxide                                                                                |
| FTO             | : Fluorine doped tin oxide                                                                                    |
| ITO             | : Indium tin oxide                                                                                            |
| PCE             | : Power conversion efficiency                                                                                 |
| V <sub>oc</sub> | : Open circuit voltage                                                                                        |
| J <sub>sc</sub> | : Short circuit current                                                                                       |
| FF              | : Fill factor                                                                                                 |
| J-V             | : Current density - voltage                                                                                   |
| eV              | : Electron volt                                                                                               |
| CBM             | : Conduction band minimum                                                                                     |
| VBM             | : Valence band maximum                                                                                        |
| UV              | : Ultra violet                                                                                                |
| SEM             | : Scanning electron microscopy                                                                                |
| AFM             | : Atomic force microscopy                                                                                     |
| XRD             | : X-ray diffraction                                                                                           |
| PL              | : Photoluminescence                                                                                           |
| IPCE            | : Internal photon conversion efficiency                                                                       |
| MA              | : Methylammonium                                                                                              |
| FA              | : Formamidinium                                                                                               |
| DMF             | : N,N-dimethylformamide                                                                                       |
| DMSO            | : Dimethyl sulfoxide                                                                                          |
| ACN             | : Acetonitrile                                                                                                |
| CB              | : Chlorobenzene                                                                                               |
| t-BP            | : 4-tert-Butylpyridine                                                                                        |
| RPM             | : Revolution per minute                                                                                       |
| TiAcac          | : titanium diisopropoxide bis(acetylacetonate)                                                                |
| CHMAI           | : Cyclohexylmethylammonium iodide                                                                             |
| Li[TFSI]        | : Lithium bis(trifluoromethanesulfonyl)imide                                                                  |
| NMP             | : N-Methyl-2-pyrrolidone                                                                                      |
| PFNBr           | : Poly(9,9-bis(3'-(N,N-dimethyl)-N-ethylammonium-propyl-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene))dibromide |
| TOP $\alpha$ -3 | : ((E,E,E,E)-1,2,4,5-Tetrakis[4-[bis(4-methoxyphenyl)amino]styryl]benzene)                                    |