# Impact of Air Processing on the Performance of Perovskite Solar Cells Prepared by Sequential Solution Deposition

Capstone Report

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**Abstract:**

The transition to renewable energy has driven significant interest in solar technologies, with perovskite solar cells (PSCs) emerging as a upcoming alternative because of their potential for low-cost, high-efficiency energy conversion. However, challenges related to fabrication processes, environmental sensitivity, and scalability hinder their widespread commercialization. This capstone project investigates the effect of air-based fabrication on PSC performance, aiming to explore cost-effective, scalable production methods. Devices using Spiro-OMeTAD as the hole transport layer were fabricated under both controlled glovebox and ambient air conditions, with and without the use of self-assembled monolayers (SAMs). The impact of fabrication conditions on surface morphology was analyzed using Atomic Force Microscopy (AFM), revealing a direct correlation between smoother surfaces and improved device performance. The results show that air-processed PSCs can achieve competitive efficiencies (up to 18.1%), and SAM treatment significantly enhances performance, reaching up to 20.7% PCE. These findings highlight the viability of low-cost, air-based processing for scalable perovskite solar cell manufacturing, paving the way for future advancements in renewable energy technologies.

# Contents

# Preface

The transition to renewable energy is crucial in addressing global challenges such as climate change and resource depletion. Solar energy, with its abundance and potential for low-cost energy conversion, is at the forefront of this shift. Perovskite solar cells (PSCs) have gained attention for their high efficiency and simple fabrication processes, making them a promising alternative to traditional silicon-based solar cells. However, challenges remain in the fabrication and scalability of PSCs, as traditional methods often require inert atmospheres to prevent degradation. This capstone project explores the potential of fabricating PSCs in ambient air as a more scalable, cost-effective solution. By examining the role of Spiro-OMeTAD and self-assembled monolayers (SAMs), this study investigates how processing conditions impact device performance and surface morphology.

I would like to extend my heartfelt gratitude to my supervisor, Professor Annie Ng, whose unwavering support and guidance have been invaluable throughout this research journey. Despite the challenges of remote collaboration, your dedication and expertise have greatly enriched the quality of this work. I am also deeply grateful to the faculty and staff at Nazarbayev University, particularly the C4.134 lab team, for providing access to high-quality materials and cutting-edge facilities that made this project possible. Lastly, I appreciate the NU Library for granting free access to an extensive collection of academic resources, which was critical for the development of this research. Without the support of these individuals and institutions, this project would not have reached its full potential.

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# Chapter 1

# Introduction

## 1.1 Background

### 1.1.1 Renewable Energy and Solar Cells

The shift towards renewable energy sources has become important because of a fast-increasing global demand for sources of energy and the growing environmental concerns related to fossil fuel consumption. Among various green energy technologies, sources of solar energy are prominent due to its rich sunlight and the possibility of using it as a clean energy source. Solar energy transfer, which includes the converting of light into electricity, is achieved through photovoltaic (PV) systems, mostly known as solar cells [1]. Solar panels work by making use of the photovoltaic effect, an operation where some materials produce electrical current while they are lit. This process happens at the atomic level, where photons from the light source cause electrons to move within the material, by which electricity is generated [2]. In current days, solar devices are extremely progressed and adapted to different areas of science and technology very quickly. Starting from early silicon-based structures to last and complex perovskite solar cells, which are showing considerable effectiveness and low price [3].

### 1.1.2 Evolution of Solar Cell Technologies

We can divide the path of solar cell development into three main generations. Early solar cells, primarily built from crystalline silicon, have become the most commercially widespread due to their strong long-term stability and relatively high efficiency, generally ranging between 15% and 20% [4]. However, silicon solar cells are mainly known for their high material costs and power-consuming fabrication processes [5]. Thin-film photovoltaic mechanisms use materials such as cadmium telluride (CdTe) and copper indium gallium selenide (CIGS) that are mostly associated with a second generation of the solar cell industry. Materials that

are utilized in these solar cell manufacturers are applied in extremely thin layers onto various substrates, which allows for greater flexibility and lower material usage rather than normal silicon-based cells. This generation of solar cells is well-known for its potential to lower production expenses and be used in different areas of technology, such as creating integrated photovoltaics [6]. These solar panels are cheaper to build and they provide greater elasticity for various applications due to their thin-film component. However, their energy efficiency is comparatively smaller than conventional silicon-based solar cells, limiting their total performance at some conditions [7].

The last generation of solar cells, the most latest advancement in photovoltaic technology, combines a wide diapason of innovative materials and complex technology structures that target to solve the problems faced by earlier generations, such as limited efficiency, high production costs, and scalability issues. Perovskite solar cells are the promising technology of nowadays generation of solar cells due its lower cost and higher energy efficiency. Discovered in 2009, perovskite materials, which are defined by their unique crystalline structure ($ABX_3$), have undergone rapid grow, with their power conversion efficiency (PCE) starting of 3% at their inception to over 25% within just a decade. This fast development not only introduces perovskite solar cells as one of the most efficient types of photovoltaic devices but also ranks them as a notable competitive variant in the global solar energy market. Their size at the global demand, low-cost fabrication processes (such as solution preparation), and possibility for coming into flexible and lightweight devices have made them a main advantage in both academic and industrial research, further underlining their future to takeover the renewable energy area [8]. This research project will talk about specifically perovskite solar cells made by sequential solution deposition method, and the complex structure of these cells, study the role of self-assembly materials in enhancing their performance.

### 1.1.3 Perovskite Solar Cells

Perovskite solar cells are named after the perovskite structure, which is constructed by simple chemical structure of $ABX_3$ (as shown in Figure 1.1). In this structure, "A" and "B" are cations of different sizes, with "A" typically being the larger cation. The "X" represents an anion, commonly a halide such as iodide, bromide, or chloride. This exceptional position of ions establishes a crystalline structure that provides perovskite materials their unique ability to absorb light and transport energy, making them extremely effective for solar devices. In the terms of solar panels, the usual perovskites that used are hybrid organic-inorganic lead halides, such as methylammonium lead iodide ($MAPbI_3$). These type of materials have differential electronic features, such as good light absorption abilities, extended carrier diffusion lengths, and controllable band gaps. These characteristics make them

to efficiently use sunlight and transform it into electricity, making them highly effective for photovoltaic practice [8].

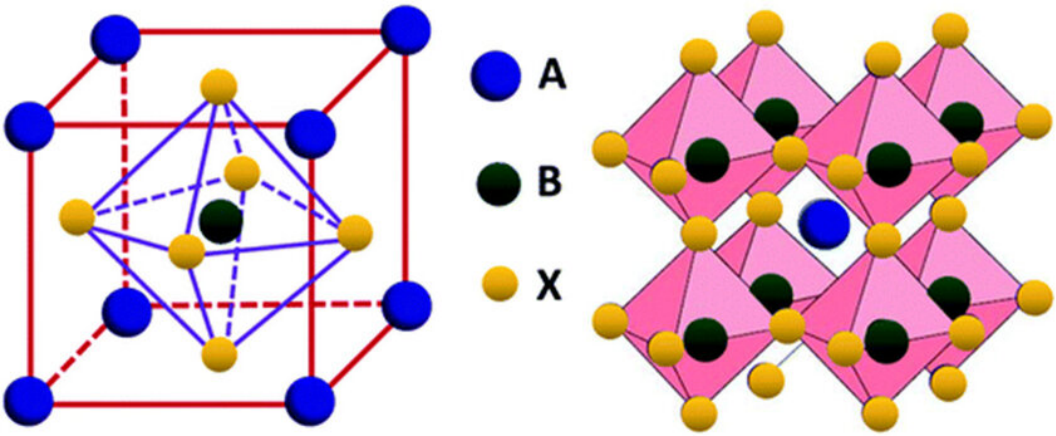


**Figure 1.1:** Perovskite chemical structure

The perovskite solar cells are very easy to establish and operate. They can be manufactured using solution-based methods at relatively low temperatures, which radically decreases production costs compared to silicon-based solar cells [8]. Furthermore, perovskite materials are adaptable to different envireonments and can be deposited on a variety of substrates. For instance, flexible plastics, permitting the enhancement of lightweight, flexible solar panels [9]. This positive side of the device is useful in different areas where electronics or photovoltaics are mostly utilized.

On the other side, perovskite solar cells also encounter huge challenges. The main one is the stability of such devices. Especially, at the normal environmental conditions it will probably will take place. [10, 11]. Researchers are studying and operating seduously to address these issues, and one of the most promising ideas implicates developing the architecture of the solar cell itself, leading to the manufacturing of perovskite solar cells prepared by sequential solution deposition [12].

### 1.1.4 Sequential Deposition Techniques in Perovskite Thin Film Fabrication

The two-step sequential deposition technique is a cornerstone in the fabrication of high-efficiency perovskite solar cells (PSCs), particularly when aiming for precise control over film crystallinity, morphology, and interface quality. Unlike the one-step method—where precursor solutions are mixed and deposited simultaneously—the two-step method decouples the deposition of metal halide (e.g., $PbI_2$)

and the organic halide (e.g., MAI or FAI). This procedural separation offers enhanced control over reaction kinetics, grain size, and the elimination of undesirable intermediate phases.

A major benefit of the two-step method lies in its facilitation of large grain formation and reduced trap density, which are vital for efficient charge transport and minimized recombination losses. Liu et al. [13] demonstrated that introducing ascorbic acid during this process creates a porous $PbI_2$ layer that enhances infiltration of the organic salt, resulting in a pure perovskite phase and high device stability. Similarly, Zheng et al. [14] reported that mitigating interfacial tensile stress using LiOTf at the $SnO_2/PbI_2$ interface allows for controlled $PbI_2$ nucleation, yielding high-quality films with a PCE of 25.33%. Chen et al. [15] further improved upon the method by integrating a "quasi-2D" perovskite template, which induced vertical grain orientation during crystallization. This approach suppressed the formation of non-photoactive $\delta$-phases and produced devices with remarkable efficiencies up to 25.79%, along with long-term operational stability.

For mixed tin-lead perovskites, which are inherently more sensitive to environmental degradation, the two-step method proves advantageous. By tailoring the crystallization conditions and employing additives like MOFs (metal-organic frameworks), researchers have mitigated oxidation issues and achieved improved film uniformity and lifetimes [16]. Yin et al. showed that MOF (UiO-66) integration in $FASnI_3$ films improved PCE and stability over 100 days under inert storage conditions. Despite its promise, the two-step method is not without challenges. A significant issue is the incomplete conversion of $PbI_2$, which can leave residual lead halide, degrading the interface and reducing device performance. However, optimization strategies involving additive engineering, humidity tuning, and post-annealing treatments continue to resolve these shortcomings. Feng et al. [17] revealed that halide additives in the perovskite precursor promote larger crystal growth and defect passivation, further enhancing both efficiency and stability in blade-coated films.

In summary, the two-step deposition method stands as a robust and versatile approach for perovskite solar cell fabrication. Its adaptability across diverse perovskite chemistries, coupled with its compatibility with ambient and scalable processing, makes it an essential tool in the future of high-performance, commercially viable photovoltaics.

### 1.1.5 Self-Assembly Materials in Enhancing Perovskite Solar Cells

Self-assembly means the operation by which molecules spontaneously construct themselves into precise, stable design without the need for outside interference [18]. In perovskite solar cell design, self-assembly materials are crucial in improving the quality of crystal structure, making it more easy for eletrons and holes to

move across different layers, and decraesing the number of defects. These effects are important to achieve improved efficiency and increased stability in solar cells [19].

The main way to develop the working quality of perovskite solar cells is by usage of self-assembled monolayers (SAMs). These are very thin layers of organic molecules that mainly form one, organic layer on a surface of the substrate. When SAMs are allocated between the perovskite and charge transport layers, they help construction of energy levels, decrease energy losses, and improves the overall effectiveness of the solar panels [19]. Moreover, self-assembly processes can be used to create ordered nanostructures within the perovskite layer itself, which can also develop light absorption and charge transport. These nanostructures allow for more efficient production of photogenerated carriers, reducing potential recombination and enhancing the energy transfer of the solar cells [20].

### 1.1.6 Future Directions

Despite remarkable advancements in laboratory-scale perovskite solar cells (PSCs), their large-scale commercialization faces several persistent challenges. Key obstacles include environmental stability, toxic lead content, interface degradation, and variability in performance under ambient conditions. These issues must be addressed through coordinated materials engineering, scalable processing techniques, and improved encapsulation strategies. One promising area is the development of ambient-processable PSCs. Lang et al. [21] reported a molecular interaction strategy enabling high-efficiency (23.72%) methylammonium-free perovskites printed under ambient air, retaining over 92% of performance after 672 hours at 85 °C. This progress is vital for enabling roll-to-roll manufacturing of flexible modules, reducing energy costs in device fabrication.

Another critical direction is improving the mechanical and environmental robustness of flexible PSCs. Tang et al. [22] highlighted how innovations in transparent electrodes, flexible substrates, and hybrid perovskite formulations have pushed flexible PSC efficiencies beyond 25%. Continued research into these components can expand the use of PSCs into wearable electronics, aerospace, and building-integrated photovoltaics (BIPV). From a materials standpoint, novel approaches for stabilizing tin-based and lead-free perovskites are essential for reducing environmental impact. Yin et al. [16] demonstrated that integrating UiO-66, a zirconium-based metal-organic framework (MOF), significantly enhanced the crystallinity and lifetime of tin halide PSCs under inert storage, marking an important step toward sustainable solar technologies.

Interface engineering, particularly through the use of self-assembled monolayers (SAMs), remains a rich domain for innovation. SAMs like Br-4Pacz have shown substantial potential in passivating interfacial trap states and aligning energy lev-

els. Recent reviews [23] emphasize the need for robust, thermally stable SAMs that function across diverse fabrication environments, particularly in air-processed and tandem configurations. In this context, exploring the combined effects of air processing and surface modification strategies becomes crucial. This capstone project, titled *"Impact of Air Processing on the Performance of Perovskite Solar Cells Prepared by Sequential Solution Deposition,"* seeks to directly address this intersection. By examining how ambient fabrication conditions influence perovskite morphology and photovoltaic behavior—particularly when stabilized by interfacial SAMs such as Br-4Pacz—this research contributes to bridging the gap between laboratory protocols and scalable manufacturing practices. It also offers insights into optimizing sequential deposition methods for realistic environmental processing conditions.

## 1.2 Ethical and Professional Responsibilities

- **Ethical Responsibility:**
  The ethical responsibility of manufacturing perovskite solar cells prepared by two-step method fabrication mainly comes from materials that were used during fabrication, especially the combination of lead-based compounds. It is obvious that perovskite has high effectiveness and cheap, but usage of lead has some negative effects to the environment. Lead is a toxic heavy metal. It should be properly handled or disposed of, otherwise, it can contaminate water and soil, causing serious health issues for humans and damaging the environment. For that reason, from ethical perspective it is important to ensure that manufacturing process of the cell structure of the device is followed the proper safety protocols. It can contain different methods for the safe recycling of perovskite solar cells that includes lead compounds. Also, there will be other methods such as replacing lead-based perovskite solar cells with lead-free perovskites to decrease the risks.

  The accessibility of the technology might be another ethical concern. Perovskite solar cells can make solar energy more cheaper, but it's important to make sure that everyone has equal access to the positive sides of the perovskite technology. It may contain considering how these solar cells can be made affordable and accessible to low-income communities and developing nations where clean energy is most needed. By solving these ethical problems, the whole plan of the research can focus on developing main energy aspects of the solar cells industry.

- **Informed Judgments:**
  Looking for the growing path of the perovskite solar cells it is important to make next step of the device development by considering both technical and societal aspects of the knowledge. Starting from technical side, the materials

that were used in manufacturing, design of the device itself, and experiments that are meant to test the device should be made through good research. This includes staying updated on the latest advancements in self-assembly materials and interface engineering.

From societal point of view, actions of research must also take into account the broader implications of the technology. For example, discussions around supply chain ethics, specifically about choosing raw materials for the device, should be a factor in decision-making. Most of the time materials used in solar cells are metals and rare earth elements. There will be ethical issues such as human rights violations and environmental degradation considering the regions of mining where those elements extracted. Guaranteeing that these materials are ethically found and that the project follows fair trade activities is important for responsible development.

By weighing both the technical performance and societal impact of each decision, the project can maintain a balance between innovation and social responsibility.

- **Global Context:**
  The advances and design of perovskite solar cells fabricated by sequential solution deposition are important overall. In areas with significant sunlight, particularly in developing countries, perovskite solar panels may provide a solution to energy problems. The lower manufacturing costs and flexibility of these cells compared to traditional silicon cells may facilitate the wider adoption of solar energy in parts of the world where initial investment in energy infrastructure is a barrier.

  However, it is also important to consider that different regions of the world may have varying regulatory frameworks for the use of materials like lead. While some nations can be forced to keep strong rules on the usage of hazardous materials, others can not have good infrastructure and management to impose those rules. For that reason, the worldwide manufacture of those devices should be regulated by locals to ensure safety and service.

- **Economic Impact:**
  The economic effect of this project covers both small and long-term effects. In an unsustained period, the production of perovskite solar cells with self-assembly materials could stimulate economic activity by creating jobs in the research, development, and manufacturing sectors. Given that perovskite solar cells are cheaper to manufacture compared to regular silicon-based solar cells, the commercialization of this technology could also reduce the total expenditure for solar energy, making it more available to consumers and businesses alike.

In the long term, the widespread acceptance of perovskite solar cells can change the energy issue by decreasing the cost of green energy, which could result in significant savings for both consumers and governments. Moreover, the integration of self-assembly materials could increase the durability and efficiency of solar cells, leading to longer-lasting devices and reduced maintenance costs.

However, there are possible economic issues as well. The original investment in increasing fabrication processes and installing recycling systems for the dangerous materials used in these cells could be expensive. Also, instability in the availability and price of raw materials, such as lead or other perovskite materials, can enhance production costs.

- **Environmental Impact:**
  One of the most crucial discussions of this research project is its environmental impact. Sources of solar energy are much more sustainable than fossil fuels, however, the materials used in perovskite solar cells, particularly lead-based compounds, have some serious environmental concerns. If it is not properly managed, lead contamination from perovskite cells can cause long-term environmental damage. Thus, one of the main environmental responsibilities of this research project is to find lead-free alternatives or implement safe recycling practices for end-of-life solar cells.

  In contrast, using self-assembly materials opens up the possibility of making these solar cells more environmentally friendly. These materials can improve the efficiency of solar cells, which means more energy can be generated with a lower number of resources. Furthermore, self-assembled materials are often more energy-efficient to build, which could decrease the overall carbon mark of the production process.

  By solving these environmental problems and encouraging sustainable processes, this research project can make way for the development of solar devices that not only can give clean energy but also decrease environmental harm.

- **Societal Impact:**
  The public effect of this research project is huge, particularly in terms of improving the approach to cheap and sustainable energy. In nations where electricity is critical or expensive, the progress of low-cost, efficient solar cell devices can give people a secure and safe source of energy, contributing to the developed quality of technology and economic development. For example, rural areas in developing countries, where traditional energy infrastructure is lacking, could benefit greatly from decentralized solar energy systems powered by perovskite solar cells.

Furthermore, the conversion to sources of renewable energy has a greater public impact, including the decrease of air pollution and the softening of climate change, which directly affect public health and well-being. By helping to a cleaner energy future, this research project aligns with global efforts to develop more sustainable societies.

Additionally, the commercialization of perovskite solar cells could reduce energy differences by making solar devices more accessible to poor populations. By guaranteeing that the positives of this technology are distributed fairly, this research project has the future to enable communities and advance social justice in the conversion to green energy.

# Chapter 2

# Methodology

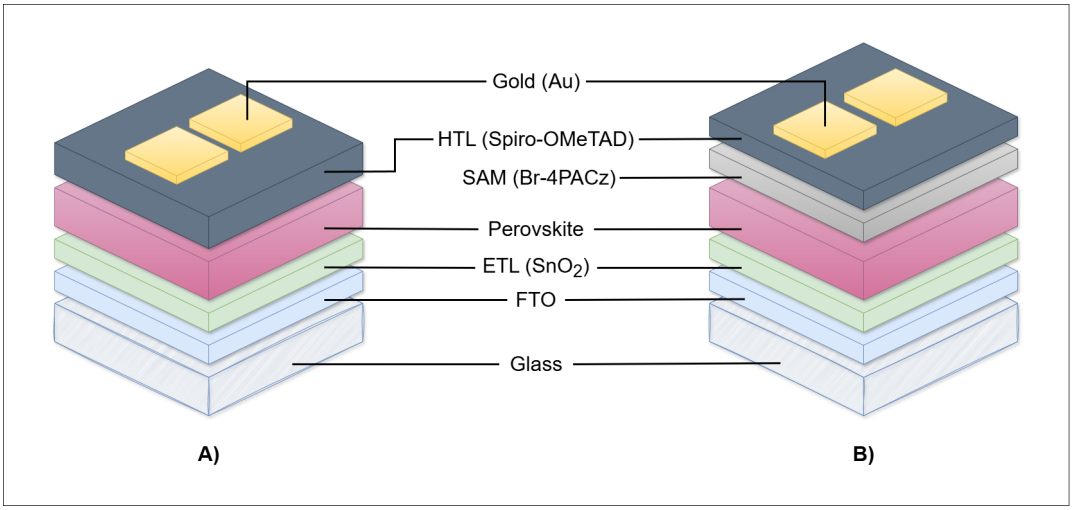


**Figure 2.1:** Device configuration of perovskite solar cells A) with only Spiro-OMeTAD layer; B) with SAM added layer

## 2.1 Materials

The materials used in this study were carefully selected to ensure high reproducibility, stability, and performance in the fabrication of perovskite solar cells (PSCs). Transparent fluorine-doped tin oxide (FTO) coated glass substrates served as the foundational conductive layer, chosen for their low sheet resistance and high optical transmittance. As the electron transport material, a colloidal dispersion of tin oxide ($SnO_2$) nanoparticles was utilized due to its excellent electron mobility, optical clarity, and favorable energy level alignment with the conduction band of the perovskite absorber. All chemicals were handled in clean laboratory conditions,

and reagents were used as received from suppliers without further purification unless otherwise noted.

The perovskite light-absorbing layer was fabricated using a sequential deposition method involving two precursor solutions. The first contained inorganic salts—lead iodide ($PbI_2$) and cesium iodide (CsI)— were dissolved in dimethylformamide (DMF), forming the metal halide foundation of the perovskite film. The second solution, comprising formamidinium iodide (FAI), methylammonium bromide (MABr), and methylammonium chloride (MACl) in isopropanol (IPA), provided the organic cations and halides needed to complete the perovskite crystal formation. In select devices, a self-assembled monolayer (SAM) of Br-4Pacz was introduced between the perovskite and hole transport layers to reduce interfacial defects and improve energy alignment. Spiro-OMeTAD, doped with lithium bis(trifluoromethanesulfonyl)imide (Li-TFSI) and 4-tert-butylpyridine (TBP), functioned as the hole transport layer in all device architectures. All solvents, including acetonitrile and chlorobenzene, were anhydrous grade and sourced alongside the chemical precursors from established vendors such as Sigma-Aldrich, Alfa Aesar, and Greatcell Solar.

## 2.2 Perovskite Solar Cell Fabrication

All perovskite solar cells (PSCs) were fabricated using a planar n–i–p heterojunction architecture via a two-step sequential solution deposition technique. To investigate the influence of ambient environmental conditions and interfacial engineering on device performance, four fabrication pathways were systematically explored, differing in processing atmosphere and hole transport layer (HTL) composition.

### A. Substrate Preparation

Commercial fluorine-doped tin oxide (FTO) glass substrates (sheet resistance $\approx$ 15 $\Omega$/sq) were used as transparent bottom electrodes. Substrates were etched using zinc powder and hydrochloric acid to define active areas. They were then sequentially ultrasonicated in detergent, deionized water, acetone, and isopropanol (15 minutes each), followed by nitrogen drying and 15 minutes of UV-ozone treatment to enhance surface wettability.

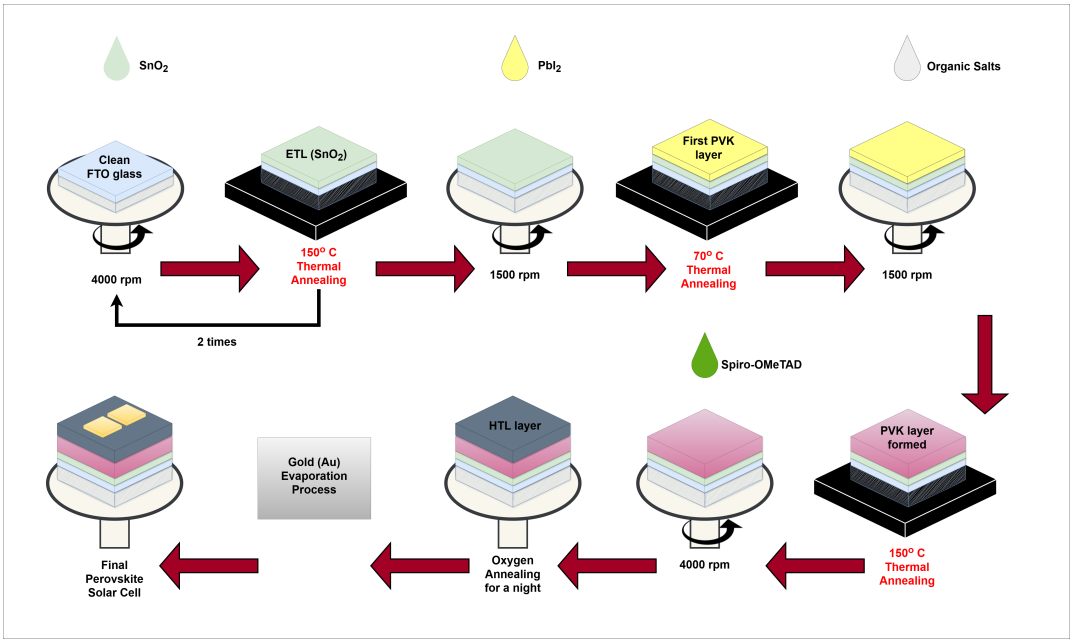


**Figure 2.2:** Device fabrication process of perovskite solar cell

### B. Electron Transport Layer (ETL)

A tin oxide ($SnO_2$) nanoparticle dispersion (15 wt%, Alfa Aesar) was diluted with deionized water in a 1:3 volume ratio and filtered (0.45 µm PTFE). The solution was spin-coated at 4000 rpm for 30 s and annealed at 150 °C for 30 min in ambient air. After cooling down to room temperature we repeated the step.

### C. Perovskite Absorber Layer

The perovskite layer was fabricated using a two-step method. A solution of $PbI_2$ (1 M) and CsI (1 M) in DMF:DMSO (95:5 v/v) was first spin-coated at 2000 rpm for 30 s. This was followed by spin-coating an organic halide solution of formamidinium iodide (FAI), methylammonium bromide (MABr), and methylammonium chloride (MACl) in isopropanol (60, 6, and 6 mg/mL, respectively) at 4000 rpm for 30 s. Films were annealed at 135 °C for 15 min to complete crystallization.

### D. Interfacial SAM Modification

For Condition 4 devices, a self-assembled monolayer (Br-4Pacz) was applied after perovskite formation. The SAM was deposited via dynamic spin-coating at 4000 rpm for 30 s with no thermal annealing.

### E. Hole Transport Layer (HTL)

Spiro-OMeTAD (72.3 mg/mL in chlorobenzene) was doped with 30 µL of 4-tert-butylpyridine (TBP) and 35 µL of Li-TFSI solution (260 mg/mL in acetonitrile) per 1 mL. The solution was filtered (0.22 µm PTFE) and spin-coated at 4000 rpm for 30 s.

### F. Electrode Deposition

Gold (Au) electrodes ($\sim$100 nm) were deposited via thermal evaporation under high vacuum ($\sim 10^{-6}$ Torr) using a shadow mask to define an active area of 0.06 $cm^2$.

### G. Fabrication Configurations

- **Condition 1 (Control):** $PbI_2$ and $SnO_2$ layers were deposited in a glovebox, while organic halide deposition and annealing were done in air. HTL: Spiro-OMeTAD only.
- **Condition 2 (Fully Air-Processed, Spiro Only):** All steps were performed in ambient air (30–40% RH), without SAM.
- **Condition 3 (Fully Glovebox-Processed, Spiro Only):** All fabrication steps were completed in a nitrogen-filled glovebox. No SAM applied.
- **Condition 4 (Fully Air-Processed, SAM + Spiro):** All steps were done in air. SAM (Br-4Pacz) was deposited before Spiro-OMeTAD.

## 2.3 Characterization

To assess the performance and structural quality of the fabricated perovskite solar cells, two core characterization techniques were employed: current–voltage (J–V) measurements to evaluate photovoltaic efficiency and atomic force microscopy (AFM) to analyze surface roughness of perovskite films and complete devices.

### Photovoltaic Efficiency Measurement

The photovoltaic performance of each device configuration was evaluated through current density–voltage (J–V) measurements under simulated sunlight conditions. A calibrated AM1.5G solar simulator (100 $mW/cm^2$) was used to replicate standard test illumination, and measurements were conducted using a Keithley 2400 source meter. The scan was performed in both forward and reverse bias directions to check for hysteresis effects. The key photovoltaic parameters extracted included:

- Open-circuit voltage ($V_{OC}$)

- Short-circuit current density ($J_{SC}$)
- Fill factor (FF)
- Power conversion efficiency (PCE)

All devices were tested under identical conditions, and multiple devices were measured per condition to ensure statistical reliability. The performance metrics were used to compare the effects of fabrication environment (air vs. glovebox) and the incorporation of a self-assembled monolayer (Br-4Pacz) on overall device efficiency.

### Surface Roughness Characterization

Atomic Force Microscopy (AFM) was utilized to explore the surface morphology and roughness of both perovskite layer (after deposition and annealing) and the completed device stack (after HTL application). Scans were conducted using a SmartSPM 1000 system operating in tapping mode over a 5 µm $\times$ 5 µm area. The root mean square (RMS) roughness values were extracted to quantify surface uniformity, crystal growth quality, and the influence of fabrication conditions on morphological features. These measurements were critical for correlating film quality with device performance under different processing routes. ‘

# Chapter 3

# Results and Discussions

## 3.1 Photovoltaic Performance Analysis

This study evaluated the photovoltaic behavior of perovskite solar cells fabricated under distinct environmental conditions with and without self-assembled monolayer (SAM) enhancement. The core analysis focused on three configurations using only Spiro-OMeTAD as the hole transport layer (HTL): a control device (partial glovebox-air), a fully air-processed device, and a fully glovebox-fabricated reference. These variants serve as benchmarks to understand the practical limitations and potentials of air-based perovskite processing.

### Performance of Spiro-Only Devices

The **control device**, fabricated with partial glovebox exposure, delivered PCEs of 17.1% (reverse) and 18.0% (forward). These values represent a realistic performance baseline, reflecting typical laboratory workflows where perovskite films are exposed to ambient air during key steps. The fill factors (78.48% and 80.57%) indicate relatively good interfacial quality despite environmental exposure.

The **fully glovebox-processed device** achieved the highest $V_{OC}$ (1.03 V) and FF (77.08%), resulting in superior PCEs of 19.6% and 19.7%. These improvements are attributed to uniform crystallization and minimal oxygen/moisture interference, confirming the advantages of an inert environment for film integrity and interface optimization.

In contrast, the **fully air-processed Spiro-only device** showed a moderate drop in performance. $V_{OC}$ fell to 0.88 V (reverse) and 0.91 V (forward), while FF declined slightly to 78.12% and 79.77%, producing PCEs of 17.2% and 18.1%. Despite reduced interfacial quality, these results are impressive and indicate that bulk perovskite properties are largely retained even under ambient processing.

**Table 3.1:** Photovoltaic Parameters of Perovskite Solar Cells Under Different Fabrication Conditions

| **Device Configuration** | **Scan Direction** | $V_{OC}$ (V) | $J_{SC}$ (mA/cm$^2$) | FF (%) | PCE (%) |
|---|---|---|---|---|---|
| Control (Spiro-OMeTAD) | Reverse | 0.88 | 24.81 | 78.48 | 17.1 $\pm$ 0.3 |
| | Forward | 0.90 | 24.82 | 80.57 | 18.0 $\pm$ 0.2 |
| Glovebox ($*$) | Reverse | 1.02 | 24.84 | 77.08 | 19.6 $\pm$ 0.2 |
| | Forward | 1.03 | 24.82 | 76.49 | 19.7 $\pm$ 0.3 |
| Air-Processed ($**$) | Reverse | 0.88 | 24.86 | 78.12 | 17.2 $\pm$ 0.4 |
| | Forward | 0.91 | 24.88 | 79.77 | 18.1 $\pm$ 0.3 |
| SAM (0.5 mg) + Spiro-OMeTAD | Reverse | 1.10 | 25.84 | 72.69 | 20.7 $\pm$ 0.3 |
| | Forward | 1.08 | 25.85 | 67.59 | 18.8 $\pm$ 0.4 |
| SAM (1.0 mg) + Spiro-OMeTAD | Reverse | 1.09 | 25.53 | 73.66 | 20.4 $\pm$ 0.2 |
| | Forward | 1.04 | 25.49 | 69.62 | 18.5 $\pm$ 0.3 |
| SAM (1.5 mg) + Spiro-OMeTAD | Reverse | 1.08 | 25.02 | 72.22 | 19.5 $\pm$ 0.3 |
| | Forward | 1.04 | 24.98 | 69.13 | 18.0 $\pm$ 0.2 |

## Comparison to SAM-Enhanced Devices

Devices incorporating Br-4Pacz SAMs reached a peak PCE of 20.7% (reverse scan, 0.5 mg), with $V_{OC}$ up to 1.10 V and $J_{SC}$ up to 25.85 mA/cm$^2$. These enhancements are attributed to passivation of surface trap states and improved energy level alignment at the HTL interface. However, higher SAM concentrations (1.0 mg and 1.5 mg) showed reduced fill factor and PCE, likely due to excessive interface modification impeding charge transport.

Although SAM-enhanced devices demonstrated the highest absolute efficiencies, Spiro-only devices remain more relevant to industrial scale-up due to their simplicity, material compatibility, and ease of processing.

## Air Effects and Fabrication Viability

Among the Spiro-only devices, the largest environmental impact was observed in the open-circuit voltage, which ranged between 0.88 V and 1.03 V. This trend reflects increased trap-assisted recombination and poorer energy level alignment at the HTL interface in air-processed cells. Nonetheless, fill factors and $J_{SC}$ remained fairly stable across fabrication environments, indicating preserved charge transport and minimal bulk degradation.

These findings suggest that **air processing affects interfacial quality more than bulk material properties**. With optimized deposition steps and humidity control, functional PSCs with PCE $>$ 18% can be fabricated reliably under ambient conditions.

**Perovskite solar cells can be reliably fabricated in air without major efficiency losses**, especially when using Spiro-OMeTAD as the HTL and maintaining procedural consistency. While glovebox processing provides optimal performance, the results show that ambient fabrication is highly viable. **Furthermore, the addi-**

**tion of SAM layers such as Br-4Pacz can enhance the efficiency of air-fabricated devices**, offering an effective strategy for future scalable, high-performance perovskite solar technologies.

## 3.2 Surface Morphology and Roughness Analysis (AFM)

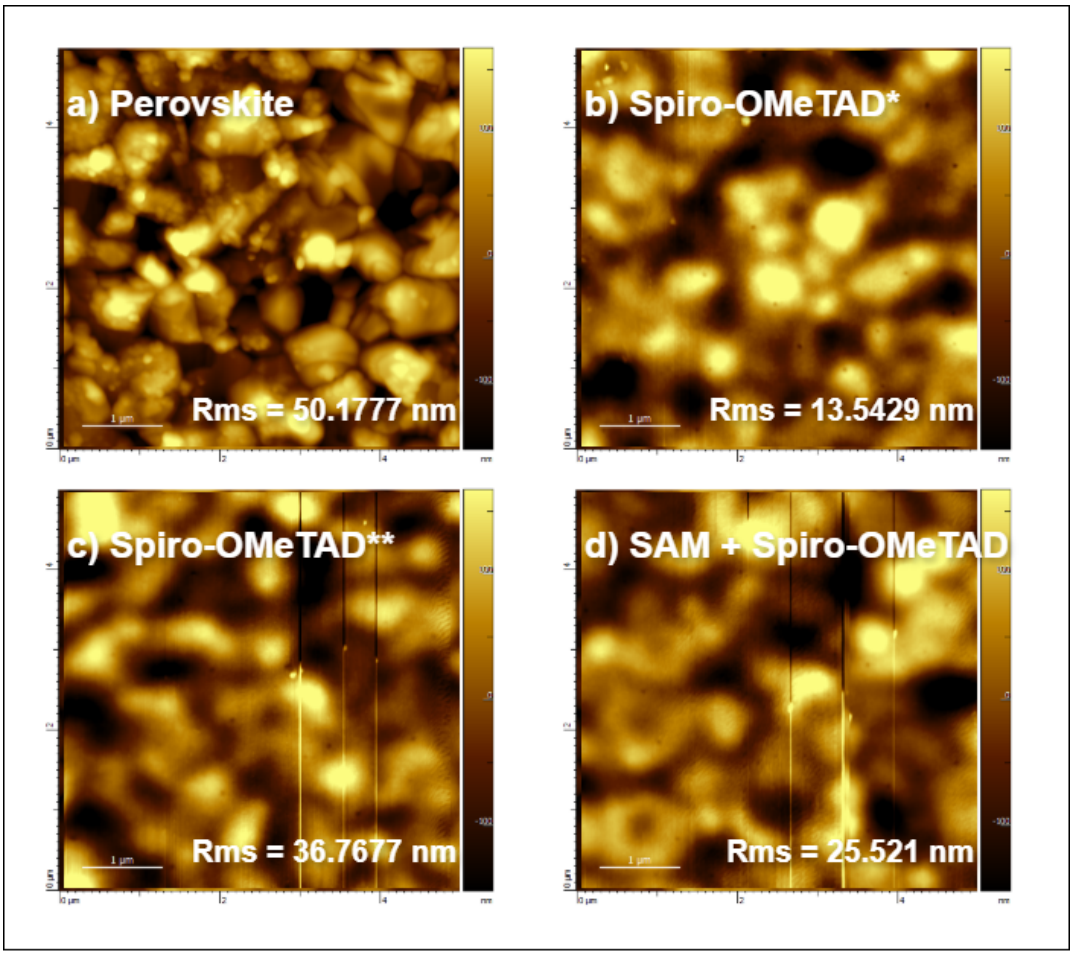


**Figure 3.1:** AFM images of a) Perovskite; b) Spiro-OMeTAD*; c) Spiro-OMeTAD**; d) SAM + Spiro-OMeTAD

Atomic Force Microscopy (AFM) was employed to examine the surface morphology and roughness of the perovskite films and the completed devices. The AFM measurements were conducted in tapping mode over a 5 $\mu$m $\times$ 5 $\mu$m scan area, providing high-resolution topographical data. The **perovskite films** (before Spiro-OMeTAD deposition) exhibited the highest roughness, with Ra = 40.44 nm and RMS = 50.18 nm (as shown in Figure 3.1 a)). This rough surface likely contributed

to higher interfacial recombination losses and reduced hole extraction efficiency, impacting the overall device performance.

The **Spiro-OMeTAD layer processed entirely in a glovebox** showed significantly smoother surface characteristics, with Ra = 11.03 nm and RMS = 13.54 nm (as shown in Figure 3.1 b)). This smoother surface allowed for more efficient charge transport and hole extraction, contributing to the higher performance of these devices, which achieved the best efficiency (PCE of 19.7%). The reduced roughness in glovebox-processed devices indicates that a controlled atmosphere minimizes surface defects and ensures better interfacial quality.

In contrast, the **Spiro-OMeTAD layer processed entirely in air** exhibited much higher roughness, with Ra = 30.73 nm and RMS = 36.77 nm (as shown in Figure 3.1 c)). The increased roughness, likely due to uncontrolled crystallization in ambient conditions, resulted in higher interfacial resistance and reduced efficiency (PCE of 17.2%). Interestingly, the **SAM-modified Spiro-OMeTAD devices** (with 0.5 mg Br-4Pacz) showed intermediate roughness (Ra = 21.37 nm, RMS = 25.52 nm, as shown in Figure 3.1 d)) and improved performance (PCE of 20.7%), highlighting the potential of SAM treatment to reduce surface roughness and enhance device efficiency, even under air-processing conditions.

# Chapter 4

# Conclusion

In this capstone project, the impact of air processing on the performance of perovskite solar cells (PSCs) was thoroughly explored, with particular emphasis on the role of surface morphology and interfacial engineering. Through a series of controlled experiments, it was demonstrated that **perovskite solar cells can be successfully fabricated in ambient air without major downsides**, provided that careful attention is given to processing conditions and film uniformity. Devices fabricated entirely in air, using only Spiro-OMeTAD as the hole transport layer (HTL), achieved competitive efficiencies of up to **18.1%**, demonstrating the feasibility of air-based fabrication for PSCs.

The **glovebox-processed devices** consistently outperformed their air-processed counterparts, with PCEs exceeding **19.7%**. This improvement was attributed to the **controlled environment** that minimized moisture and oxygen exposure, leading to better perovskite crystallization and more stable interfaces. However, **air-processed devices** still showed promising efficiency, highlighting the potential for **low-cost, scalable production** of high-performance PSCs without the need for inert atmospheres.

Further enhancements in efficiency were observed when **self-assembled monolayers (SAMs)** such as **Br-4Pacz** were introduced. SAM treatment significantly improved **device performance**, particularly under air-processing conditions, by reducing interfacial defects and improving hole extraction at the perovskite/HTL interface. The highest efficiency (20.7%) was achieved with **0.5 mg of Br-4Pacz**, suggesting that **SAM treatment** can serve as a viable method for improving the **performance of air-fabricated PSCs**.

Through surface morphology analysis using Atomic Force Microscopy (AFM), it was established that **smoother perovskite films** correlate directly with higher photovoltaic efficiency. Devices fabricated in the **glovebox environment** exhibited the smoothest surfaces, leading to reduced recombination losses and enhanced charge transport. Even in air-processed devices, **SAM modification** improved sur-

face quality, further enhancing performance.

In conclusion, this study demonstrates that **air-based processing** can serve as a viable route for fabricating **high-performance perovskite solar cells**, with **SAM treatment** offering a straightforward method to improve efficiency, even in ambient conditions. These findings lay the groundwork for future research focused on **optimizing fabrication techniques** for **cost-effective, scalable production** of **next-generation solar technologies**.

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