ADVANCING PEROVSKITE SOLAR CELL FABRICATION THROUGH LARGE-AREA ATMOSPHERIC PLASMA PROCESSING

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Abstract

This work demonstrates the use of diffuse coplanar surface barrier discharge (DCSBD) plasma as a low-temperature, solvent-free method for scalable fabrication of perovskite solar cells (PSCs). Operating below 70 °C and at atmospheric pressure, DCSBD plasma enables rapid sintering of TiO_2 and SnO_2 electron transport layers (ETLs), activation of indium tin oxide (ITO) electrodes, and surface passivation of $MAPbI_3$ perovskites. These treatments improve crystallinity, wettability, and charge transport, achieving efficiencies comparable to conventional methods while reducing energy demand. The approach offers a sustainable route for roll-to-roll production of flexible PSCs.

Keywords: Perovskite solar cells (PSCs); diffuse coplanar surface barrier discharge (DCSBD); atmospheric plasma; low-temperature processing; TiO_2; SnO_2; ITO; $MAPbI_3$; roll-to-roll fabrication

1. INTRODUCTION

The transition from conventional silicon-based photovoltaics to novel thin-film and hybrid solar technologies represents a key step toward sustainable, low-cost energy generation [1, 2]. Among the emerging alternatives, perovskite solar cells (PSCs) have attracted significant attention due to their exceptional power conversion efficiencies, facile tunability, and compatibility with flexible and printable substrates [3, 4]. However, despite the rapid laboratory-scale progress, the scalability and industrial feasibility of PSC fabrication remain hindered by the limitations of conventional processing methods, particularly those requiring high temperatures, vacuum conditions, and complex chemical treatments [5, 6]. These factors not only increase production costs but also restrict the choice of substrates, limiting the potential for roll-to-roll (R2R) manufacturing on flexible and eco-friendly materials [5].

In the pursuit of scalable and sustainable PSC production, atmospheric-pressure plasma technologies have emerged as a promising alternative for material modification, surface activation, and thin-film processing under mild thermal conditions. Plasma-based methods enable precise surface engineering without the need for solvents or high-temperature annealing, offering significant environmental and economic advantages [7–10]. In particular, diffuse coplanar surface barrier discharge (DCSBD) plasma has shown unique potential for large-area, homogeneous treatment of functional layers used in perovskite devices. Operating at atmospheric pressure and temperatures below 70 °C, DCSBD plasma delivers a high-power density and reactive environment capable of inducing physicochemical transformations in nanostructured materials while preserving the integrity of temperature-sensitive substrates such as PET, PEN, or cellulose-based nano-paper [11].

Our recent studies have demonstrated that atmospheric plasma treatment can enhance the wettability, crystallinity, and electronic properties of various semiconducting and dielectric materials, including graphene oxide, titanium dioxide, tin oxide, graphitic carbon nitride, and MXenes. These effects are critical for optimizing charge transport, interfacial adhesion, and overall device performance in PSCs. By tailoring plasma exposure time and gas composition, it becomes possible to achieve targeted surface functionalization suitable for different layers of perovskite devices, such as transparent electrodes, electron transport layers (ETLs), hole transport layers (HTLs), and perovskite absorber films themselves.

In this work, we present how plasma fabrication of perovskite solar cells, focusing on the integration of DCSBD plasma treatments into scalable, low-temperature manufacturing workflows. Specifically, we investigate plasma-assisted approaches for (i) low-temperature sintering of mesoporous TiO_2 and SnO_2 ETLs using DCSBD plasma to achieve full mineralization and crystallization below 70 °C; (ii) rapid activation and surface cleaning of indium tin oxide (ITO) electrodes in ambient air, replacing multi-step solvent cleaning; and (iii) controlled surface modification of the $MAPbI_3$ perovskite absorber, enabling defect passivation and energy-level tuning for improved charge extraction and long-term stability. The results highlight how diffuse atmospheric plasma can replace conventional high-temperature or wet-chemical steps, thus paving the way toward rapid, environmentally benign, and industrially viable PSC production compatible with flexible substrates and continuous R2R fabrication.

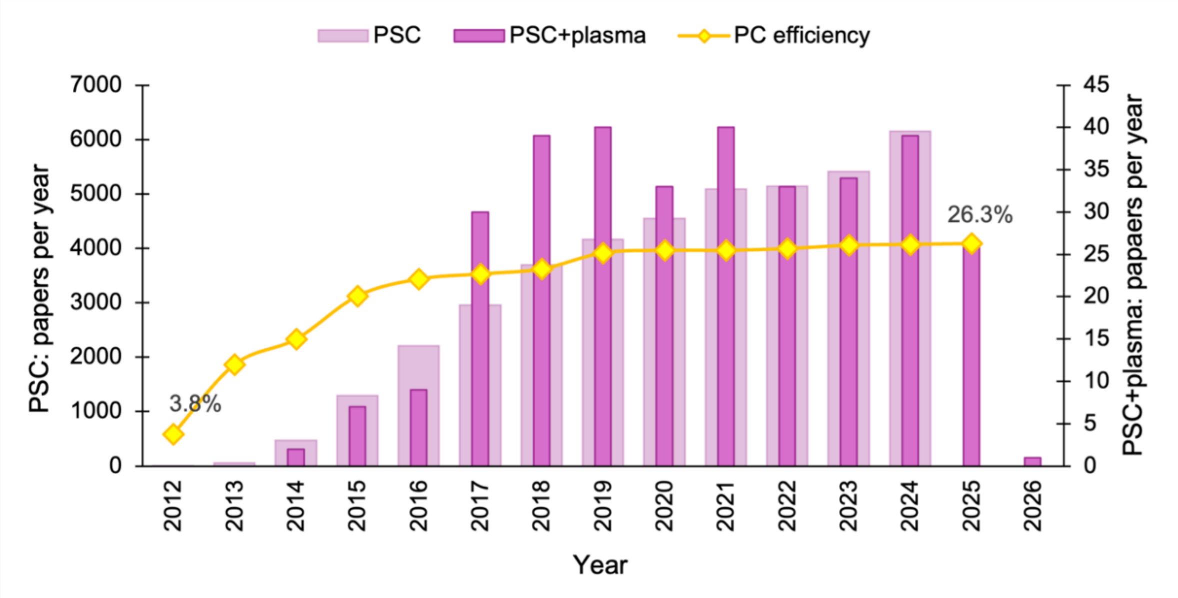
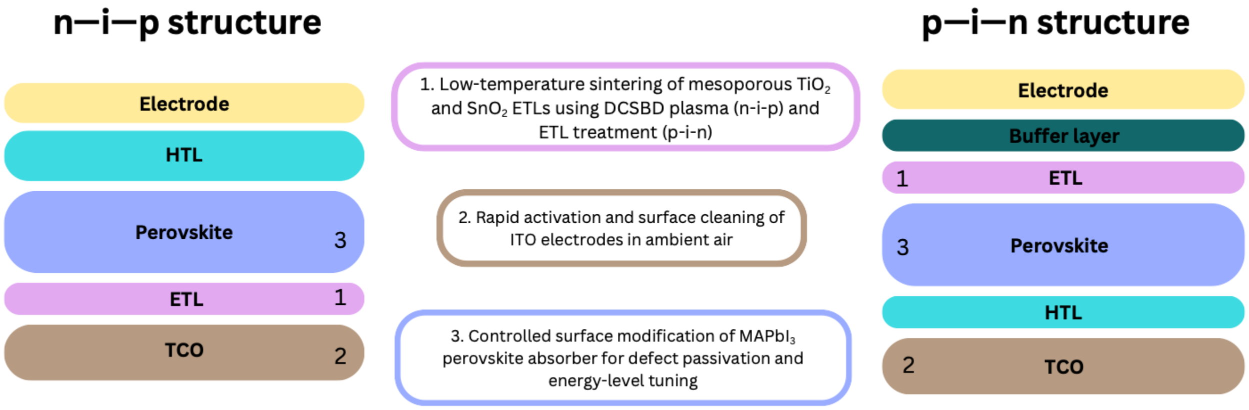


Figure 1 Annual number of publications related to perovskite solar cells (PSC) and PSC+plasma treatments, shown as bar charts (left and right axes, respectively), plotted together with the record power conversion efficiency of perovskite solar cells per year (line, right axis). Data on efficiencies were extracted from the NREL Best Research-Cell Efficiency Chart (accessed November 2025).



1. Low-temperature sintering of mesoporous TiO_2 and SnO_2 ETLs using DCSBD plasma (n-i-p) and ETL treatment (p-i-n)

2. Rapid activation and surface cleaning of ITO electrodes in ambient air

3. Controlled surface modification of $MAPbI_3$ perovskite absorber for defect passivation and energy-level tuning

Figure 2 Plasma-assisted processes integrated into perovskite solar-cell fabrication.

2. EXPERIMENTAL

2.1 Plasma Treatment Setup

All plasma treatments were performed using a DCSBD plasma source operated at atmospheric pressure. The discharge electrode consists of parallel embedded copper strips covered by a dielectric ceramic layer, generating a homogeneous, high-power diffuse plasma at a frequency of 15–25 kHz and power density up to 100 W·cm^{-3}. The plasma was sustained in various working gases (air, N_2, Ar) at temperatures below 70 °C, ensuring compatibility with thermally sensitive materials. Samples were placed approximately 0.3 mm above the plasma surface, and treatment times ranged from 1 to 10 seconds, depending on the material and process step (ITO activation, ETL sintering, or perovskite surface modification).

2.2 Fabrication of Perovskite Solar Cells

Two device architectures were fabricated: n–i–p and p–i–n.

- For the n–i–p configuration, fluorine-doped tin oxide (FTO) substrates were coated with a compact TiO_2 blocking layer followed by a mesoporous TiO_2 (m-TiO_2) or SnO_2 ETL. These layers were either thermally sintered or plasma-processed. The active layer consisted of a methylammonium lead iodide ($MAPbI_3$) perovskite, followed by Spiro-OMeTAD as the HTL and a gold electrode.
- For the p–i–n configuration, ITO served as the transparent electrode. After plasma activation, a PEDOT:PSS hole transport layer was deposited, followed by $MAPbI_3$, a PCBM electron extraction layer, and an aluminum back contact.

Plasma treatment was applied selectively to specific layers: ITO, TiO_2, SnO_2, or $MAPbI_3$, prior to subsequent deposition steps to improve surface energy, purity, and charge transport properties.

3. RESULTS AND DISCUSSION

3.1 Plasma-Assisted Low-Temperature Sintering of Mesoporous TiO_2 Electron Transport Layers

The development of efficient ETLs remains a cornerstone in advancing the performance and scalability of PSCs. Among available semiconductors, TiO_2 is the most commonlyused ETL due to its suitable band alignment, high electron mobility, and chemical stability. However, conventional sintering of TiO_2 nanostructures typically requires temperatures about 400 °C for extended times (30–60 min), which excludes its use on flexible or temperature-sensitive substrates and is incompatible with roll-to-roll (R2R) fabrication. To overcome this bottleneck, plasma-assisted mineralization represents an innovative route for rapid, low-temperature consolidation of TiO_2 films, eliminating the need for energy-intensive thermal treatment. Our team demonstrated a fast and low-temperature approach for the mineralization of inkjet-printed mesoporous TiO_2 photoanodes using DCSBD plasma operating in ambient air at approximately 70 °C. The method enabled the oxidation and removal of organic residues from a methyl-silica binder incorporated in TiO_2/silica hybrid coatings, effectively transforming the printed films into photoactive mesoporous structures. The plasma, characterized by a high power density (~100 W·cm^{-3}) and uniform discharge over large areas, provided strong oxidative species (mainly O and N radicals) capable of decomposing organic moieties within seconds while maintaining the structural integrity of TiO_2 nanoparticles. Electrochemical and photocatalytic characterizations revealed that plasma-treated layers exhibited a significant enhancement in photocurrent response and photoactivity. Linear sweep voltammetry and chronoamperometry showed a monotonic increase in photocurrent density with plasma exposure time, indicating improved charge transport and reduced recombination due to the removal of insulating organic residues. Likewise, photocatalytic tests using coumarin as a fluorescent probe confirmed that oxidative plasma treatment enhanced the generation rate of hydroxyl radicals, particularly in thinner and more porous films where plasma penetration was more effective.

These plasma-treated mesoporous coatings were subsequently implemented as ETLs in n–i–p PSCs to evaluate their photovoltaic performance. Remarkably, devices incorporating DCSBD plasma-sintered TiO_2 layers exhibited power conversion efficiencies (PCEs) comparable to those fabricated with conventionally sintered TiO_2 at 400 °C for 1 h, confirming that plasma processing can fully replace high-temperature annealing. The plasma-based approach thus enables a rapid, energy-efficient, and substrate-friendly fabrication route while maintaining the structural integrity and electronic quality essential for high-performance perovskite devices.

Building upon the earlier work on proprietary TiO_2/silica hybrid layers, where plasma enabled low-temperature mineralization of mesoporous films, the approach was extended to commercial TiO_2 nanoparticle pastes (30NR-D, Dyesol) to assess its impact directly in n–i–p PSCs. Using DCSBD plasma operated in ambient air at ~70 °C, mesoporous TiO_2 layers were sintered within minutes, achieving full oxidation of organic binders and restoration of stoichiometric TiO_2 comparable to conventional 400–500 °C thermal processing.

When incorporated into complete FTO/c-TiO_2/m-TiO_2/$MAPbI_3$/Spiro-OMeTAD/Au devices, the plasma-treated ETLs exhibited power conversion efficiencies (PCEs) equivalent to thermally sintered references, confirming that plasma exposure ensured sufficient conductivity, crystallinity, and interfacial quality for efficient charge transport. The optimized plasma treatment (2–3 min) yielded stable device operation with PCEs above 15%, high open-circuit voltage, and improved fill factor, reflecting reduced transport resistance and enhanced carrier lifetime. These results clearly demonstrate that ambient-air DCSBD plasma sintering can replace traditional furnace-based annealing, significantly reducing energy consumption and processing time. The method preserves perovskite morphology, ensures efficient electron extraction, and expands PSC fabrication toward low-temperature, scalable, and flexible photovoltaic technologies.

3.2 Plasma-Assisted Low-Temperature Sintering of SnO_2 Electron Transport Layers

Following the successful plasma sintering of mesoporous TiO_2, the approach was extended to planar p–i–n perovskite solar cells employing SnO_2 as the ETL. SnO_2 is an attractive alternative to TiO_2 due to its excellent conduction band alignment with perovskite absorbers, high electron mobility, and chemical stability; however, its processing typically requires annealing above 150 °C, limiting compatibility with flexible substrates. Using DCSBD plasma operated in argon at atmospheric pressure, highly crystalline and uniform SnO_2 layers were prepared within 5 minutes at temperatures below 60 °C, achieving structural and electronic properties equivalent to films annealed at 180 °C.

The plasma treatment efficiently removed residual organics, increased surface hydroxylation, and preserved stoichiometric SnO_2, resulting in smooth, pinhole-free layers with improved interfacial quality. When integrated into p–i–n device architectures (ITO/SnO_2/$MAPbI_3$/Spiro-OMeTAD/Au), the plasma-treated ETLs delivered PCEs of 15.2%, comparable to thermally annealed references, while exhibiting higher current densities and superior operational stability under ambient conditions. These findings confirm that atmospheric DCSBD plasma enables a fast, low-temperature, and scalable route to fabricate high-quality SnO_2 ETLs for p–i–n perovskite solar cells, advancing the development of flexible and R2R photovoltaic technologies.

3.3 Plasma-assisted Activation of Indium Tin Oxide Electrodes

Proper preparation of the ITO electrode is critical for efficient PSCs. Conventional solvent-based cleaning and UV–ozone treatment effectively remove contaminants but require several processing steps and extended times, making them unsuitable for large-scale or flexible manufacturing. In contrast, atmospheric plasma activation provides a rapid, dry, and low-temperature alternative, achieving full surface cleaning and activation of ITO in as little as 2 seconds at temperatures below 70 °C. The plasma removes carbon contamination more efficiently than chemical methods (from ~22 at.% to <5 at.%), increases hydroxyl group density, and improves wettability (contact angle <10°) without affecting morphology or conductivity.

Perovskite solar cells fabricated on plasma-treated ITO electrodes exhibited the same or slightly higher power conversion efficiencies as those prepared by conventional chemical cleaning, while the plasma process reduced preparation time from minutes to seconds. This demonstrates that plasma activation is a faster, cleaner, and fully scalable alternative for ITO treatment, ideally suited for continuous roll-to-roll manufacturing of high-efficiency PSCs.

3.4 Plasma-assisted treatment of the Methylammonium Lead Iodide ($MAPbI_3$) Perovskite Layer

The plasma modification of the perovskite absorber represents the final step in PSCs. In another work brief atmospheric plasma exposure (1–8 s, <60 °C, N_2 environment) was shown to fine-tune the surface chemistry of $MAPbI_3$ without damaging its crystal structure. The plasma introduced energetic electrons, ions, and radicals that fragmented surface organics and slightly disturbed the equilibrium of ionic vacancies (MA^+, Pb^{2+}, I^-), leading to a Pb-enriched, defect-passivated surface. This mild etching removed ~2 nm of organic residues and suppressed oxidation by reducing Pb=O species. These atomic-scale rearrangements shifted the valence and conduction bands upward, improving charge extraction and reducing recombination losses. When integrated into inverted PSCs (ITO/PEDOT:PSS/$MAPbI_3$/PCBM/Al), 2 s plasma treatment enhanced short-circuit current and open-circuit voltage, yielding PCE ≈ 11.7%, compared to 10.3% for untreated films. Devices retained >94% efficiency after 35 days, demonstrating both performance and stability gains. In summary, ultrashort plasma exposure effectively cleans, passivates, and tunes the surface energetics of perovskites through controlled chemical reactions of reactive plasma species, offering a scalable, non-destructive tool for roll-to-roll PSC fabrication.

4. CONCLUSION

We demonstrate the versatility of DCSBD plasma as a rapid, low-temperature, and solvent-free method for fabricating perovskite solar cells (PSCs). Operating at atmospheric pressure and below 70 °C, DCSBD plasma provides a homogeneous flux of reactive species capable of modifying diverse interfaces crucial to device performance.

Across all layers of the PSC architecture, plasma treatment replaced conventional thermal or chemical steps: it activated ITO electrodes, sintered mesoporous TiO_2 and SnO_2 ETLs, and engineered $MAPbI_3$ perovskite surfaces. These processes improved surface cleanliness, crystallinity, wettability, and band alignment, resulting in enhanced charge transport and stability. By addressing every key interface—electrode/ETL, ETL/perovskite, and perovskite/HTL—the DCSBD plasma approach enables a unified, energy-efficient, and scalable route to perovskite device manufacturing. Its atmospheric operation and compatibility with flexible substrates make it a promising technology for roll-to-roll production of next-generation photovoltaics.

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