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2D Modification for Humidity Resistance and Performance Enhancement in Perovskite Solar Cells

Hungyu Zhu

BASIS International School Guangzhou, Guangzhou, China
a3091841458@gmail.com

Abstract. Perovskite solar cells (PSCs) have high power conversion efficiency (PCE) and low fabrication cost. However, they are sensitive to moisture, which limits their commercial use. This study innovatively investigates a two-dimensional (2D) capping layer integrated onto a three-dimensional (3D) perovskite structure to enhance both photovoltaic performance and humidity resistance. The 2D capping layer was formed by the reaction of 2-(2-aminoethyl) benzenethiol(AEBT) with excess PbI_2. The 2D-modified perovskite films exhibited greater hydrophobicity, as showed by a contact angle of 51°, compared to 33° for the normal perovskite film, suggesting better moisture resistance. Nonetheless, the 2D-modified perovskite films showed enhanced device performance. Due to its improved crystallinity, the 2D-modified perovskite devices demonstrated higher open-circuit voltage (V_{OC}), short-circuit current density (J_{SC}), fill factor (FF), and PCE, and their narrower statistical distributions indicating improved reproducibility. The best performing 2D-modified device outperformed the best performing normal device in J-V characteristics, indicating enhanced charge transport and efficiency. Higher hydrophobicity helps reduce moisture damage in perovskite films, which is key for making PSCs more commercially viable.

Keywords: 2D/3D hybrid perovskite solar cells, Stability, Power conversion efficiency, Humidity Resistance

1. Introduction

Photovoltaic technology (PV) is key for sustainable energy. It provides high-efficiency, low-cost solar power, which reduces global reliance on fossil fuels and mitigate climate change.. In China, Guangzhou has become a representative city actively adopting PV technology across its urban infrastructure. For instance, the Guangzhou Museum of Art integrates photovoltaic panels into its architectural design (see Figure 1a), demonstrating how cultural landmarks can embrace clean energy while maintaining aesthetic value. Meanwhile, the Guangzhou Baiyun International Airport has established a large-scale PV power station (see Figure 1b), significantly increasing the proportion of green electricity used in aviation facilities. These facilities not only highlight Guangzhou's determination to transition toward renewable energy, but also demonstrate how PV technology is key to creating green, sustainable cities.

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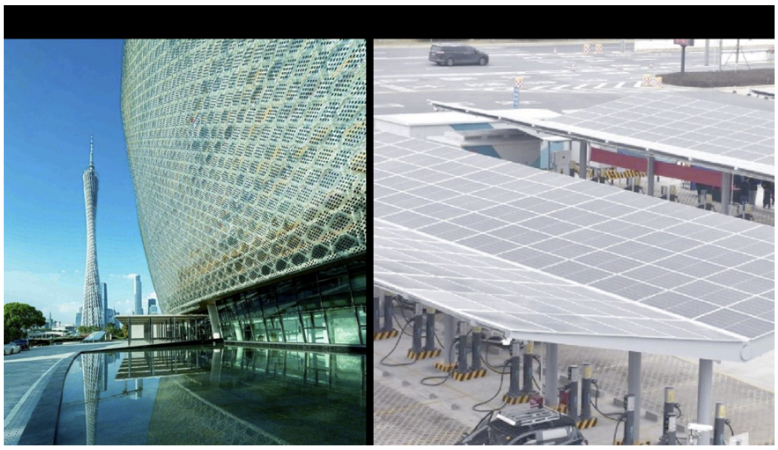

Figure 1. Application of Photovoltaic Technology in Guangzhou: (a) The Guangzhou Museum of Art integrates photovoltaic panels into its architectural design; (b) The Guangzhou Baiyun International Airport has established a large-scale PV power station

In the urban commercialization and widespread adoption of PV technology, power conversion efficiency (PCE), durability, and production costs are the main considerations. Silicon solar cells, with PCE of more than 20%, lifetime of more than 25 years, and low manufacturing costs of 0.3 $/W, account for more than 90% of the market share of the commercialized PVs [1]. Currently, the only PV that might be able to compete with silicon PV is probably the perovskite solar cells (PSCs). PSCs are hybrid organic-inorganic lead halide materials with the general formula ABX_3, where A and B are cations and X is a halide anion. These materials exhibit exceptional solar cell performance due to their high optical absorption, low exciton binding energy, high charge carrier mobility, long diffusion length, and large diffusion coefficient [2]. These advantages enable the fabrication of thin, lightweight, and potentially flexible solar cells, positioning them as a compelling alternative for scalable solar energy solutions.

In 2009, the first PSC reached a PCE of 3.8% [3]. An important breakthrough to solid-state PSC in 2012, replacing the unstable liquid electrolytes with solid-state hole transport materials, boosted PCE to around 10% [4]. Subsequent advancements resulted in rapid increase in PCE to 15.4% in 2013, 20.1% in 2015, and 25.7% by 2021, and as of July 2025, the record for single-junction PSCs is at 27%, highlighting the potential of PSCs [5].

However, despite their high PCE, instability hinders the commercialization of PSCs, particularly their sensitivity to moisture. Perovskite materials, like methylammonium lead iodide (MAPbI3), are sensitive to moisture. Humidity catalyze the irreversible decomposition of $MAPbI_3$ into aqueous HI, solid PbI_2, and CH_3NH_2 [6]. Exposure to relative humidity above 50% rapidly reduces $MAPbI_3$ solar cell performance [7]. This vulnerability poses a significant challenge for their employment in real-world environments. Thus, addressing their instability is critical to the commercialization of PSCs.

To mitigate humidity-induced degradation, 2D perovskite structures have emerged as a promising strategy. By incorporating large organic cations, 2D perovskites form layered structures that enhance moisture resistance while maintaining optoelectronic properties. This study investigates a 2D-modified PSC, where a 2D layer is applied to a 3D perovskite to improve both performance and stability (see TOC figure in Abstract). Specifically, we introduce a 2D capping layer on top of the perovskite active layer. We aim to demonstrate that 2D modification enhances cell performance and reduces the adverse effects of humidity.

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This work innovatively formed 2D capping layer by the reaction of 2-(2-aminoethyl) benzenethiol (AEBT) with excess PbI_2. AEBT served as the 2D capping material because of its special mix of functional groups and aromatic structure (see Figure 2). Amino and thiol groups reduce surface defects by coordinating with undercoordinated Pb^{2+} ions, thereby reducing non-radiative recombination. Meanwhile, the hydrophobic aromatic ring improves moisture resistance, providing a robust barrier against humidity-induced degradation. These features make AEBT an ideal 2D modifier for simultaneously enhancing the efficiency and stability of perovskite solar cells.

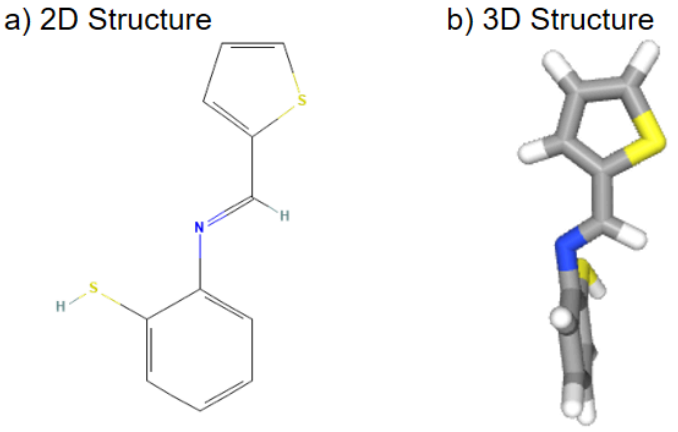

Figure 2. The 2D and 3D structure of 2-(2-aminoethyl)benzenethiol (AEBT)

2. Experiments

2.1. Materials and device fabrication

Normal Perovskite. Normal perovskite films were prepared using a one-step spin-coating method on fluorine doped tin oxide (FTO) coated glass substrates with a TiO_2 electron transport layer (ETL). A precursor solution was made by dissolving lead(II) iodide (PbI_2) and methylammonium iodide (MAI) in a stoichiometric ratio in a mixed solvent of dimethylformamide (DMF) and dimethyl sulfoxide (DMSO). The solution was spin-coated at 4500 rpm for 30 seconds onto the TiO_2-coated FTO substrate, followed by annealing at a set temperature of 120 °C for 1 min to form the 3D perovskite film. Subsequently, a Spiro-OMeTAD hole transport layer (HTL) was deposited onto the perovskite film by spin-coating, followed by thermal evaporation of a gold (Au) electrode to complete the n-i-p device structure.

2D-Modified Perovskite. For the 2D-modified perovskite, a two-step process was employed. Initially, the 3D perovskite film was prepared as described above. Subsequently, a solution of AEBT in isopropanol was spin-coated onto the 3D perovskite film at a 1500 rpm for 30 seconds. 2D perovskite layer was formed by the reaction of AEBT with excess PbI_2. The resulting films were annealed at 150 °C for a 10 min to form a 2D capping layer on the 3D perovskite structure. Similarly, the device was completed by depositing a Spiro-OMeTAD HTL and an Au electrode.

2.2. Characterization

UV-Vis Absorption Spectroscopy. UV-Vis spectroscopy is a common analytical technique for the quantitative determination of various analytes, including transition metal ions, conjugated organic molecules, and biological macromolecules. Although measurements are most commonly performed

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in solution, the method can also be employed to investigate solid and gaseous samples. In our research, absorption spectra were recorded by UV3600 at a range from 500 nm to 800 nm.

Photoluminescence (PL) Spectroscopy. PL is the light emission from any form of matter after the absorption of photons. It is one of many forms of luminescence (light emission) and is initiated by photoexcitation, that is, photons that excite electrons to a higher energy level in an atom. Photoluminescence spectroscopy is a powerful optical characterization technique widely used to probe the electronic and defect properties of semiconductors. In our research, steady-state PL spectra were carried out on FLS980 with an excitation wavelength set to be 532 nm. The scanning wavelength is set to be 700 nm to 900 nm. The collection slit is about 3 mm and the collection light path is about 90°.

X-ray Diffraction (XRD). X-ray diffraction is a nondestructive analytical technique used to determine the crystallographic structure, phase composition, and orientation of materials. When a crystalline sample is irradiated with X-rays, constructive interference occurs at specific angles according to Bragg's law, producing characteristic diffraction patterns. In our research, XRD patterns were obtained using a diffractometer with Cu Kα radiation (λ = 1.5406 Å) over a scan range of 8°~45°, and the scan speed is about 0.2°/min.

Scanning Electron Microscopy (SEM). SEM is a type of electron microscopy that produces high-resolution images of a sample surface by scanning it with a focused beam of electrons. The incident electrons hit atoms in the specimen. This interaction creates signals like secondary electrons and backscattered electrons, and these signals provide detailed information about surface topography, grain structure, and film uniformity. In this study, SEM was employed to analyze the morphology of perovskite films, focusing on grain size distribution, surface smoothness, and the presence of pinholes or cracks. These factors are key for charge transport and device stability. In our research, surface morphologies were imaged using a scanning electron microscope at a specified accelerating voltage of 5.0 kV.

Contact Angle Measurement. Contact angles were measured after a water droplet dropped on the surface of thin films of normal perovskite and 2D-modified perovskite have been stabilized for 10 seconds.

3. Results & discussion

3.1. Fabrication procedures illustration

Based on the materials and device fabrication in 2.1, Figure 3 illustrates the PSC fabrication procedures. Figure 3a presents the precursor solution which was made by dissolving PbI_2 and MAI in a stoichiometric ratio in a mixed solvent of DMF and DMSO. Initially, the precursor solution appeared as a yellow gel-like substance. Figure 3b shows the 3D perovskite film. The precursor solution with TiO2-coated FTO substrate, followed by annealing to form the 3D perovskite film. Upon annealing, the 3D perovskite film formed a solid structure, accompanied by a color transition to dark red. Figure 3c demonstrates perovskite film with Spiro-OMeTAD Layer. The Spiro-OMeTAD HTL was deposited onto the perovskite film; Figure 3d illustrates Perovskite Solar Cell. We apply thermal evaporation of an Au electrode to complete the n-i-p device structure.

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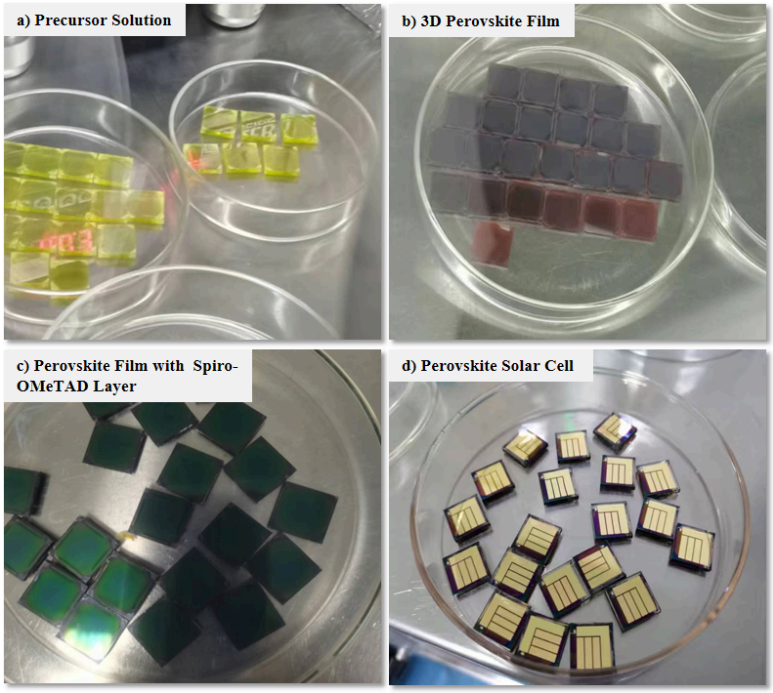

Figure 3. The perovskite fabrication process

3.2. Morphological characterization

Figure 4a shows the UV-Vis absorption spectra for both normal and 2D-modified perovskite films. The normal film shows a clear absorption profile. The onset is around 820 nm, corresponding to a bandgap of ~1.51 eV. In contrast, the 2D-modified film shows a slightly higher absorption intensity across the visible spectrum. The onset was shifted to ~780 nm, indicating a blue shift that resulted in a slightly larger bandgap of ~1.59 eV. The 2D-modified sample also shows a subtle excitonic peak at ~750 nm. This excitonic peak is a characteristic of 2D films, where quantum confinement effects lead to stronger excitonic interactions. The UV-Vis absorption spectra show that the 2D-modified film absorbs more light in the visible range, likely due to presence of the 2D layer. These optical improvements suggest that 2D modification could improve the PCE of PSCs.

Figure 4b shows the XRD patterns of the perovskite films. The normal perovskite displays a peak at ~13.3°, attributed to the (001) plane of PbI_2, alongside other characteristic perovskite peaks at 14.1° ((110) plane) and 28.4° ((220) plane). In the 2D-modified sample, the PbI_2 peak at about 13.3° is slightly weaker. Meanwhile, the other perovskite peaks are relatively sharper and more intense. The XRD patterns demonstrate that the 2D modification does not significantly affect the crystallinity of the light-absorbing 3D perovskite. The diminished PbI_2 peak at 13.3° in the 2D-modified sample indicates fewer surface defects due to PbI_2. This is important because PbI_2 can serve as a site for charge recombination. These findings are supported by Bhushan P. Kore et al., who showed that that 2D perovskite coatings on 3D perovskites contribute to reduced presence of PbI_2 [8]. The sharper and more intense perovskite peaks indicate enhanced crystallinity.

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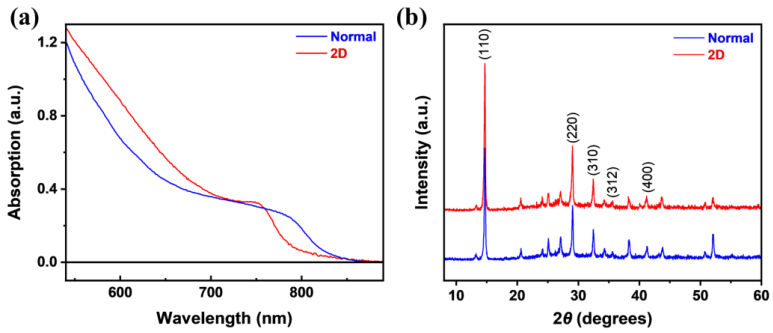

Figure 4. (a) UV-Visible absorption spectra and (b) XRD patterns of normal (blue) and 2D-modified (red) perovskite films

Figure 5a shows the PL spectra of both perovskite samples. Both the normal film and the 2D-modified film show emission peaks around 800 nm. This matches their band-edge emission. The 2D-modified film demonstrates a slightly higher PL intensity compared to the normal perovskite, indicating reduced non-radiative recombination. The PL spectra further support the improved optoelectronic properties of the 2D-modified film. The higher PL intensity indicates reduced non-radiative recombination. Since both samples have similar absorption at the excitation wavelength, the enhanced PL suggests fewer trap states, which leads to a higher open-circuit voltage (V_{OC}) in devices.

To further investigate carrier dynamics, TRPL measurements were performed. Figure 5b presents the TRPL decay curves for both samples. The 2D-modified film exhibits a slightly slower decay compared to the normal film. This suggests a longer carrier lifetime, likely due to reduced non-radiative recombination, complementing the PL measurements. This aligns with findings by Kore BP et al., who reported that 2D perovskite coatings on 3D perovskites lead to longer carrier lifetimes, enhancing device performance.[8] The longer carrier lifetime is expected to contribute to higher V_{OC} in solar cells, as charge carriers have a greater probability of being collected before recombining

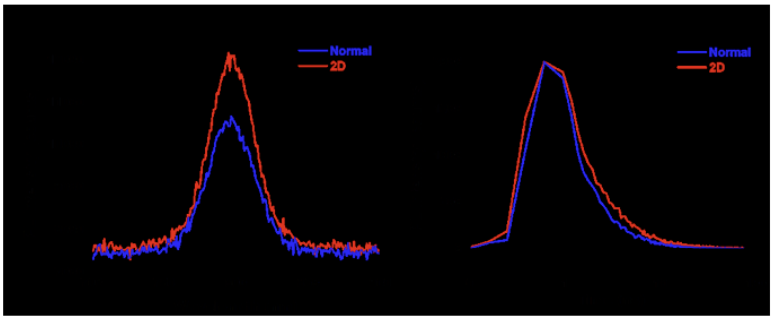

Figure 5. (a) Steady-state PL spectra and (b) TRPL decay curves of normal (blue) and 2D-modified (red) perovskite films

Figures 6a and 6b show SEM images of the normal and 2D-modified perovskite films. The normal perovskite film exhibits smaller grains with unclear grain boundaries. In contrast, the 2D-modified perovskite film has larger grains and clearer grain boundaries, suggesting improved film morphology. The SEM images further highlight the enhanced crystallinity indicated by Figure 4b. The larger grains and clearer boundaries in the 2D-modified film suggest a more uniform and compact structure. This can enhance the fill factor (FF) and overall PCE in solar cells. Larger grains

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reduce grain boundary density, minimizing recombination sites and improving charge carrier transport. The smaller gaps between grains limit moisture pathways, contributing to improved stability.

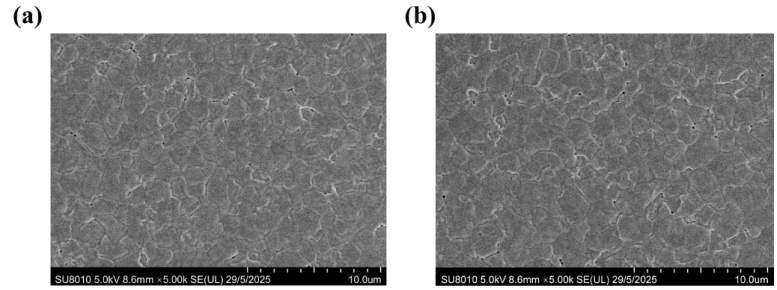

Figure 6. SEM image of the (a) normal perovskite film and (b) the 2D-modified perovskite film

3.3. Surface hydrophobicity and photovoltaic performance

Figure 7a and 7b show the contact angle measurements. The normal film surface has a contact angle of ~33°, indicating a hydrophilic nature. In contrast, the 2D-modified perovskite surface exhibits a larger contact angle of ~51°, suggesting increased hydrophobicity. Contact angle measurements demonstrate better humidity resistance in the 2D-modified film. The increased hydrophobicity reduces water adsorption and penetration. This helps reduce hydrolysis and delays degradation into volatile components.

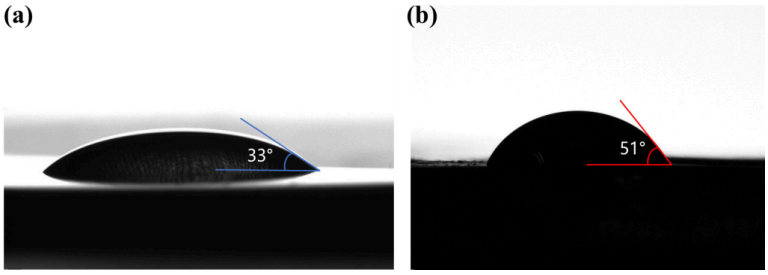

Figure 7. Contact angle measurements of the (a) normal perovskite film and (b) the 2D-modified perovskite film

Figure 8a shows the statistical distributions of V_{OC} for normal and 2D-modified PSCs. The 2D-modified PSC exhibit a higher mean V_{OC}, likely due to reduced non-radiative recombination, as indicated by the enhanced PL intensity (Figure 5a) and longer TRPL lifetimes (Figure 8b). Figure 8b depicts the statistical distributions of short-circuit current density (J_{SC}) for normal and 2D-modified PSCs. The 2D-modified PSC achieve a higher mean J_{SC}, consistent with the enhanced absorption and smaller bandgap observed in the UV-Vis spectra (Figure 4a). Figure 8c demonstrates the statistical distributions of FF for normal and 2D-modified PSCs. The 2D-modified devices show a higher mean FF, reflecting improved charge transport and reduced series resistance. The larger grain sizes (Figure 6b) and fewer grain boundaries reduce resistive losses, while the longer carrier lifetimes (Figure 5b) enhance charge collection efficiency. Figure 8d presents the statistical distributions of PCE for normal and 2D-modified PSCs. The 2D-modified devices demonstrate a higher mean PCE, reflecting the combined benefits of improved V_{OC}, J_{SC}, and FF. In all measurements, 2D-modified PSCs showed narrower statistical distributions, indicating more consistent and reproducible solar cell performance. This consistency is closely linked to fewer

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defects and uniform material quality suggested by the higher and more uniform PL intensity suggested by Figure 5a. This proves 2D modification to 3D perovskite's ability to improve cell performance.

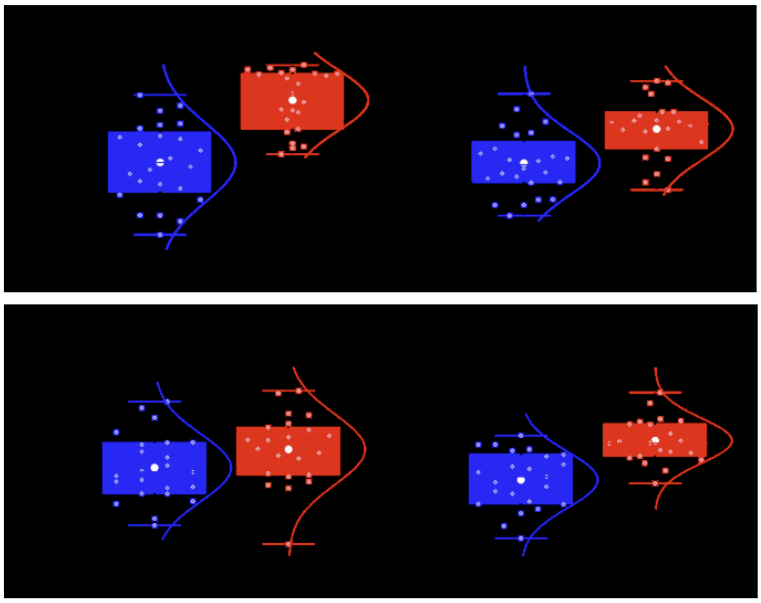

Figure 8. (a) Statistical distribution of VOC, (b) JSC, (c) PCE, and (d) FF for normal (blue) and 2D-modified perovskite (red) solar cells

Figure 9 shows the J-V curves of the best performing devices for both normal and 2D-modified PSCs. The 2D-modified device exhibits a higher V_{OC}, J_{SC}, and FF, resulting in a superior PCE compared to the normal device. The improved J-V characteristics are attributed to enhanced light absorption, reduced recombination, and better charge transport due to larger grains and fewer defects.

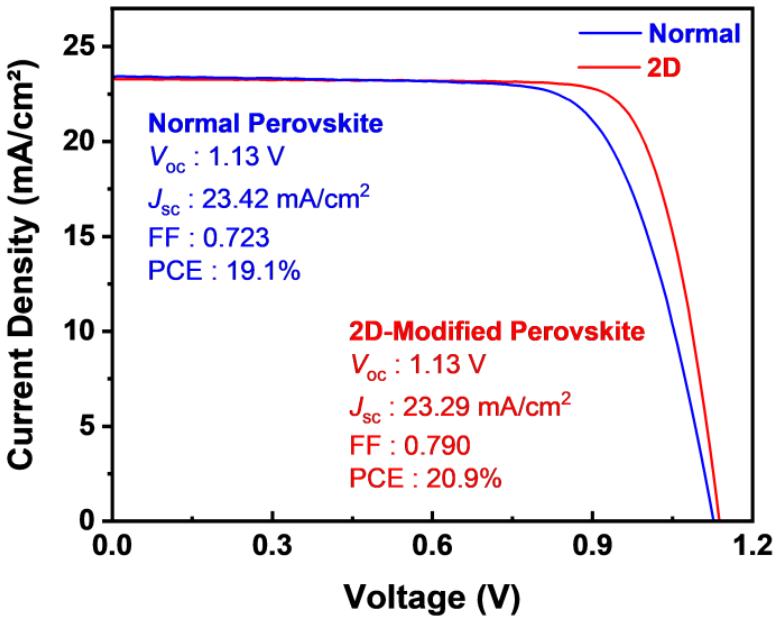

Figure 9. J-V curves of best performing device devices for normal (blue) and 2D-modified (red) PSCs

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4. Conclusions

This study shows that modifying PSCs in 2D boosts their photovoltaic performance and humidity resistance. The 2D-modified PSCs show better light absorption and a smaller bandgap. They also have less non-radiative recombination, improved crystallinity, larger grain sizes, and increased hydrophobicity. Device performance metrics show that the 2D-modified PSCs have better photovoltaic properties. This includes higher V_{OC}, J_{SC}, FF, and PCE. The J-V curves also show improved characteristics for the best performing device. The Hydrophobicity has increased, as demonstrated the increased contact angle from ~51°to ~33°. This enhanced hydrophobicity is key to reducing moisture-related damage, a major challenge towards PSC commercialization. Furthermore, the insights from this research show how 2D layers can boost stability and cell performance. This knowledge can support future material engineering efforts, potentially leading to more stable devices with better performance, thus accelerating the transition of PSCs from laboratory research to widespread commercialization. As the global demand for renewable energy continues to grow, PSCs with enhanced stability will play a pivotal role in the industry.

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