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Abstract Book of the 2024 International PhD School on

Perovskite PV

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Preface

The Abstract Book of the 2024 International PhD School on Perovskite PV encapsulates the intellectual and scholarly endeavors presented at the doctoral school held from April 15 to April 17, 2024, in Freiburg, Germany. Organized jointly by Fraunhofer Institute for Solar Energy Systems (ISE), the University of Cyprus, and Pixel Voltaic, this event served as a focal point for the comprehensive exploration of perovskite solar cells.

Throughout this gathering, PhD students, researchers, and industry players converged to delve into the intricacies of perovskite photovoltaics, addressing critical facets such as carbon-based electrodes, simulation methodologies, life-cycle assessment, tandem photovoltaics utilizing perovskite materials, industrialization hurdles, and stability assessment coupled with outdoor reliability testing.

The PhD School fostered an environment conducive to heated discussions and interdisciplinary exchange, paving the way for novel insights and collaborative initiatives in the field of renewable energy. Participants explored cutting-edge research, innovative methodologies, and emerging trends, ensuring a holistic understanding of the challenges and opportunities inherent in perovskite solar cell technology.

We extend our heartfelt gratitude to all the contributors, including the insightful speakers, tireless organizers, and dedicated participants, whose collective efforts culminated in a vibrant and intellectually enriching experience. Their invaluable contributions will undoubtedly propel the field of perovskite photovoltaics forward, accelerating the global pursuit of sustainable energy solutions.

As we embark on the journey of disseminating the knowledge and insights garnered during this event, we remain committed to fostering collaboration, innovation, and excellence in the field of perovskite solar cells.

Sincerely,

Markus Kohlstädt

Head of Team Perovskite Thin-Film Photovoltaics

Department Organic and Perovskite Photovoltaics

Fraunhofer Institute for Solar Energy Systems ISE

Table of Contents

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Two-Step Sequential Vapor-Solution Hybrid Deposition of Uniform and Stable Perovskite Films Assisted by Slot-Die Coating Intended for Commercial Large-Area Photovoltaic Applications

Ahmed Javed[1, 2, *], Mustafa Yaşa[2], Görkem Günbaş[1, 2, 3], Selçuk Yerci[1, 2, 4, *]

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Abstract

The deposition of perovskite films by slot-die coating (SDC) offers various advantages over the ubiquitously used spin coating techniques, including faster processing, compatibility with large-area substrates, less chemical consumption, cost-effectiveness, and high throughput. Moving beyond a lab-oriented spin coating technique, this research opens gates for large-area deposition of perovskite films mainly focused on commercial and industrial avenues. In this research, we have used a hybrid deposition process for the perovskite film on a 12×12 cm^2 ITO-coated glass substrate which includes both the vacuum-based and solution-based processing. A hybrid deposition of organic-inorganic mixed halide perovskite films (MA, FA, Cs) Pb $(I, Br, Cl)_3$ is achieved by thermally co-evaporating PbI_2 and CsBr followed by an SDC procedure of a mixture containing formamidinium iodide (FAI) and methylammonium chloride (MACl) dissolved in ethanol/isopropanol and butanol. The coating parameters including chuck temperature, coating speed, and pump rate were optimized on the SDC unit while the solution concentration was adjusted to form a uniform and homogenous perovskite film. The X-ray diffraction (XRD) and photoluminescence (PL) spectroscopy results confirm the existence of the α-black perovskite phase ($2\theta=14.2°$) and a bandgap of ~1.6 eV. Moreover, the uniformity of the deposited perovskite layer is depicted by a very low variance in the FWHM of both XRD and PL peaks taken throughout the large area. Furthermore, a surface compositional analysis done by X-ray photoelectron spectroscopy (XPS) featured all the characteristic elemental peaks of the perovskite composition while the absence of residual metallic Pb peak (Pb^0) showed the viability of our technique to avoid undesired decomposition of perovskite. With the power conversion efficiency of the PIN-configured SDC-processed solar cell (ITO/NiOx/Perovskite/LiF/C_{60}/BCP/Cu) comparable to the conventional spin coating technique (~18%), this industrially favorable approach can lead to a pathway toward the commercialization of perovskite solar cells.

Keywords

Perovskite, Hybrid Deposition, Slot-die Coating, Large-Area Substrates

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Monolayers with Different Anchoring Groups for Perovskite Solar Cells

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Abstract

Inverted perovskite solar cells (iPSCs) are increasingly using self-assembled monolayers (SAMs) as hole-transporting materials due to their ease of formation and cost-effectiveness. A well-known example of SAMs is phosphonic acid with a carbazole moiety and various functional groups. Despite promising results, the relationship between the molecular structure of SAMs and the electrical properties of iPSCs remains unclear. While it is well established in the literature that certain anchoring groups, like phosphonic acid, have the capability to form a monolayer with strong bonds on the surface of oxides, there is limited knowledge regarding how the anchoring group impacts the properties of the monolayers (including surface coverage, wetting angle, work function fine-tuning). However, there was a manuscript with triphenylamine-based boronic acids, where the manuscript authors claimed that boronic acids are the more suitable choice due to less acidity and potentially increased stability. Despite that, boronic acid is known to be less stable and has a weaker bond with the surface than phosphonic acid. This study aims to determine whether triphenylamine-based phosphonic acid monolayers with different functional groups are a better alternative to boronic acids for creating efficient and stable solar cells. We synthesized a series of molecules with phosphonic anchoring groups as hole-transporting materials in iPSC devices. Further research is underway to test molecules with phosphonic and boronic anchoring groups and analyze their differences to better understand how they relate to photovoltaic device performance.

Keywords

Hole Transporting Material, Self-Assembled Monolayer, Perovskite Solar Cells

Solvent Delamination for Photovoltaic Module with Polyolefin Elastomer Encapsulation Layer

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Abstract

The first step of high-quality photovoltaic (PV) module recycling is delamination, a process that aims to remove the encapsulation layer between glass and solar cells. Various methods have been investigated for the delamination of ethylene-vinyl acetate (EVA) polymer. However, the solvent delamination of polyolefin elastomer (POE) encapsulant in photovoltaics has not been described. Our research aims to present the approach of it. Its significance increases as polyolefins are expected to become the dominant polymer in PV encapsulation within the next 10 years. We used a monocrystalline glass-glass PV module with POE encapsulation layer for the research. The module was cut into samples conforming to the structure of the module. The samples were immersed into six solvents under the specified conditions. The results showed that three solvents (acetone, dimethyl sulfoxide, and isopropanol) had no effect on structural changes. Two solvents (tetrahydrofuran, and dimethylcyclohexylamine) presented swelling of POE. The solvent was absorbed by the polymer, causing an increase in the volume of samples, however, the layers remained connected. The test with ethylene glycol allowed to separate clean glass that can be used for the manufacture of new products without additional cleaning. Even though the links between glass and encapsulation layer were detached, ethylene glycol was unable to completely dissolve the polymer. The solar cells remained encapsulated in polymer on both sides. They require additional processing (e.g., thermal treatment) to remove the polymer for metal recovery from solar cells.

Keywords

Encapsulation Layer, Polyolefin Elastomer, Solvent Delamination, Recycling

Acknowledgments

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Investigation of Halide Perovskite Precursor Solutions with SAXS

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Abstract

Halide perovskites (HPs) as an absorber material for solar cells has drawn the attention of the scientific community due to their outstanding increase in efficiency up to 26.1% as well as low-cost solution-based processing methods. Despite their popularity, it is not yet fully understood how anions, cations and the solvent influence the early stages of crystallization. For this reason, we used small angle X-ray scattering (SAXS) to investigate the precursor solution of $FAPbI_3$, $MAPbI_3$, $CsPbI_3$, $RbPbI_3$, $KPbI_3$, $NaPbI_3$ and $MAPbBr_3$ in γ-butyrolactone (GBL), dimethylformamide (DMF), dimethyl sulfoxide (DMSO), N-methyl-2-pyrrolidone (NMP) and mixtures thereof. This follows a previous study where we investigated the precursor solution of $MAPbI_3$, $FAPbI_3$ and $MAPbBr_3$ in GBL, DMF and mixtures. We performed SAXS experiments at BESSY II, at PTB's four-crystal monochromator beamline using the ASAXS end station. Previously, we developed a core-shell model with $[PbX_6]$ (X = I, Br) octahedra surrounded by solvent molecules to explain the distance between adjacent scattering objects (d_{exp}). The core can be arranged as a single octahedron or as corner-sharing octahedra, depending on the anion and the solvent. It was shown that a molecular A-cation (MA^+, FA^+) does not affect the precursors' arrangement. In this study, we demonstrate that alkali metals (Na^+, K^+, Rb^+, Cs^+) as A-cation influence d_{exp} due to higher charge density than molecular cations. Thus, we extended the core-shell model assuming that the A-cation and the solvent molecules compete to surround the core. The $[PbX_6]$ octahedra are surrounded by a solvent shell with randomly oriented molecules or by an A-cation shell. The SAXS data analysis (using SASfit) shows higher polydispersity than the previous model, which indicates an increase in the heterogeneity of the solution and aligns well with the proposed extended model.

Keywords

Precursor Solution, SAXS, Halide Perovskites

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Advancing Optoelectronics: A Comparative Study on the Characterization of Perovskite Solar Cell Absorber Material with and without 2D Surface Passivation Layer Using TRMC Technique

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Abstract

The application of Time Resolved Microwave Conductivity (TRMC) as a non-contact method for assessing charge carrier kinetics in photoconductive materials has proven successful in recent studies focused on perovskite thin films. The excess charge carrier lifetimes have been measured under various conditions regarding light intensity, pulse length and excitation laser wavelength. This study demonstrates the informative value derived from TRMC measurements conducted at low laser intensities and diverse wavelengths for the evaluation of the quality of perovskite films with triple cation and triple anion compositions, using PEAI layers for surface passivation in certain cases. A strong correlation has been found between the observed changes in excess charge carrier lifetime within the absorber films and the subsequent performance of the solar cells constructed with these layers. The non-passivated sample exhibits an accelerated decay rate in comparison to its passivated counterpart in the microsecond range. Additionally, there is a significant reduction in the TRMC-amplitude in the non-passivated films relative to the passivated ones, indicating fast recombination already during the excitation.

Keywords

Perovskite Solar Cell, TRMC, Charge Carrier Life-time, Passivation

Isn’t It Ionic: Reducing Toxicity in Perovskite Precursors

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Abstract

As energy production is increasingly diversified and tailored to region and requirement, heavy interest is in photovoltaics. Part of the growing network of sustainable energy production should predictably be large scale solar harvesting, either for the grid or for off grid direct supply. However, the current marketable solar cells require a large amount of silicon purification, a resource already being heavily relied upon. There are many options for alternative active layers, but perovskite has been rapidly and competitively improving since the first use as a sensitizer. A key aspect of these cells is their solution processability, which should mean continuous printing process compatibility such as roll-to-roll slot-die coating. However, the widespread use of toxic solvents for cell synthesis could preclude them from large scale manufacturing. Less toxic solvents have been demonstrated for the Methylammonium Lead Iodide Perovskite (MAPbI) crystals, proving their viability for use in scaled-up, fully roll-to-roll coated devices. Considering the effects of polarity, hydrogen bonding and, fluctuation induced di-polarity (dispersive forces) on solubility, a fast way to adjust the solvent strength is to introduce an ionic compound into a less toxic solvent. The base solvent has a Workplace Exposure Limit (WEL) for long term working that is eight times higher (for parts per million) than the standard DMF. This study investigates the use of ionic solvents for the deposition of Formamidinium Lead Iodide (FAPbI), examining their impact on longevity and the resulting reduced toxicity in two-step deposition spin-coated devices.

Keywords

FAPbI, Ionic Liquid, Solvent, Perovskite

Understanding and Mitigating Atomic Oxygen-Induced Degradation of Perovskite Solar Cells for Near-Earth Space Applications

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Abstract

Metal halide perovskites have aroused burgeoning interest in the field of space photovoltaics owing to their versatile optoelectronic properties, high specific power potential, and extraordinary radiation tolerance. Despite the encouraging breakthroughs and extraordinary potential, the practical application of PSCs still faces the bottleneck due to different extreme conditions in space. For instance, atomic oxygen (AtOx) in low-Earth orbit is known to etch, corrode, and form metal oxide on the metal contact with PSC devices. To mitigate this issue, we report the applicability of thermally evaporated 0.7 µm silicon oxide (SiOx) encapsulation as an AtOx barrier for triple cation PSC. Moreover, the AtOx-induced degradation mechanism of phenethylammonium iodide (PEAI)-passivated and non-passivated devices are discussed and analyzed. We found that after a total exposure duration of 120 minutes, the SiOx-encapsulated cells maintained over 97% of their initial power conversion efficiency (PCE), regardless of the device type (passivated or non-passivated). In contrast, in the case of unencapsulated devices, PCEs for non-passivated and PEAI-passivated devices the PCE declined to a maximum value of 43% and 62%, respectively. In non-passivated and unencapsulated devices, AtOx has no impact on the short-circuit current density (J_{SC}) but degrades the fill factor (FF) and open circuit voltage (V_{OC}). In PEAI-passivated devices, the J_{SC} additionally degrades by almost 35%. Injection-current-dependent electroluminescence (EL) and intensity-dependent photoluminescence quantum yield (IPLQY) measurements ruled out the perovskite as the origin of PCE degradation. Instead, inefficient charge extraction and mobile ions, due to a swiftly degrading PEAI interlayer, are the primary causes of AtOx-induced device performance degradation in passivated devices, whereas a large ionic FF loss limits non-passivated devices with no change in transit time.

Keywords

2D Passivation, Atomic Oxygen, Degradation, Encapsulation, Perovskite Solar Cells

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PSC and CIGSSe Solar Cells: Merging Two Technologies in a Forward-Looking Testing Ground

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Abstract

New industrial and technological solutions for green energy and optoelectronics are needed to bypass the state of the art in methodology and performances, demanding an extra-effort from researchers in the field of material science for the synthesis, characterization, and prototyping of more performant materials and device solutions. In this framework, we show how two well-established thin-film photovoltaic technologies, perovskites and $Cu(In_{1-x}Ga_x)(S_ySe_{1-y})_2$ (CIGSSe) solar cells, can be combined to valorize their key features on the way to enhance devices conversion efficiency. Perovskite solar cells (PSCs) stand out for their ability to absorb visible light, high efficiency comparable to commercial silicon-based solar cells exceeding 25%, flexibility, semi-transparency, and lightweight properties, making them up-and-coming for various applications. Similarly, CIGSSe solar cells exhibit higher efficiency than amorphous silicon photovoltaic devices, lower toxicity, and are lighter and less rigid, making them suitable for tandem modules alongside other solar cell technologies. The CIGSSe absorber layer is a p-type material, renowned as an efficient absorber ($\alpha > 10^5$) with a tunable optical bandgap that can be adjusted within a range of 1.01 eV ($CuInSe_2$) to 2.5 eV ($CuGaS_2$) by varying the cationic (In/Ga) and anionic (S/Se) ratio of the solid solution, allowing for optimization to best match the solar spectrum, a crucial advantage in photovoltaic applications. Here we present a roadmap and the first results of our three pathways: i) Introduction of CIGSSe into PSCs: exploring the integration of CIGSSe into semi-transparent perovskite solar cells to improve their efficiency and stability; ii) CIGSSe utilization as hole transport material (HTM): investigating the potential of CIGSSe as an HTM in perovskite solar cells to enhance their performance for large-scale application; iii Tandem device CIGSSe/PSC: developing novel tandem solar cells by combining CIGSSe and PSC technologies to achieve higher overall efficiency.

Keywords

Perovskite Solar Cell, Hole Transporting Layer, CIGSSe Solar Cells, Tandem CIGSSe/PSC

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Hole Transporting Layers for Printable Perovskite Carbon Based Solar Cells

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Abstract

Since their first appearance in 2009, Perovskite solar cells (PSCs) have emerged as a highly promising solar technology, often surpassing the efficiency records of monocrystalline silicon photovoltaics. Carbon electrodes have garnered significant attention as a viable alternative to noble metal electrodes traditionally used for PSCs, due to their cost-effectiveness, stability, and scalability. Carbon-based PSCs (C-PSCs) can be processed with or without hole-selective layers (HSLs). However, for traditional HSL-free C-PSCs, the absence of an HSL has been identified as a key factor limiting their performance. The direct contact between the perovskite and the non-charge-selective carbon electrode has been recognized as a significant source of interfacial resistance loss, resulting in poor hole selectivity and recombination loss. In the literature, research studies are available concerning both organic and inorganic HTLs. In this work, both options were tested: inorganic, using CIS ($CuInS_2$), and organic, employing poly(3-hexylthiophene-2,5-diyl) (P3HT). Additionally, in the latter case, the potential of introducing a 2D material, specifically rGO (reduced graphene oxide), was also explored. The architecture of our C-PSCs includes the following layers: a glass substrate; a layer of fluorine-doped tin oxide (FTO);

an ETL; a perovskite absorber ($FAPbI_3$ formulation by air-assisted blade coating); an HTL; and a low-temperature carbon electrode. The goal of our experimentation is focused on reducing the performance gap that exists between the cells with gold counter electrodes, with which we have achieved an efficiency of 20,84%, and those with carbon electrodes.

Keywords

Perovskite Solar Cell, Carbon Electrode, Hole Transporting Layer

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Dual Laser-assisted Glass Frit Encapsulation for Efficient and Long-Term Stable N-I-P Perovskite Solar Cell

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Abstract

According to international standards (IEC 61646 climatic chamber tests), photovoltaic devices must be long-term stable under a temperature range of -40 °C to 85 °C and to a relative humidity of 85%. For this reason, perovskite solar cells (PSCs) must be manufactured from stable materials and encapsulated appropriately to avoid degradation due to moisture and oxygen exposure. Laser-assisted glass frit encapsulation has been successfully used to achieve long-term stable PSCs with n-i-p and HTM-free structures. The laser-sealing process can be achieved with a single laser beam or dual laser beams. The laser-assisted glass frit encapsulation method was initially reported for hermetically encapsulated HTM-free PSCs at *ca.* 100 °C using a single laser beam. Later, an advanced novel dual laser beam glass frit sealing process was developed and optimized to hermetically encapsulate n-i-p PSCs, allowing to decrease the process temperature to 65 ± 5 °C. The present work reports the use of advanced laser-assisted glass frit encapsulation using dual laser beams for encapsulating PSCs devices with different electron transport layers (ETL) and hole transport layers (HTL). Preliminary results of encapsulated n-i-p PSCs with poly[bis(4-phenyl)(2,4,6-trimethylphenyl) amine] (PTAA) as HTL at a process temperature of 65 ± 5 °C for 1 h, presented a reduction in average power conversion efficiency (PCE) from 14.18 ± 2.4% to 12.32 ± 0.90%. The reduction in performance was mainly due to a decrease in fill factor (FF) from 0.67 ± 0.09 to 0.56 ± 0.04. It was registered a slight decrease in open circuit voltage (V_{OC}) from 1.01 ± 0.03 V to 0.99 ± 0.02 V, while the short circuit current density (J_{SC}) increased from 20.73 ± 0.77 mA cm^{-2} to 21.97 ± 0.46 mA cm^{-2}. The performance loss of *ca.* 10% is related to the long exposure to heat during the laser sealing process.

Keywords

Hermetic Encapsulation, Dual Laser, Glass Frit, Perovskite Solar Cell

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Advanced Indoor Characterization and Degradation Analysis of Perovskite Mini-modules Using Several Optoelectronic Techniques

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Abstract

Perovskite materials have excellent prospects for semiconducting applications due to their desirable photoelectric properties. A lot of indoor and outdoor characterization testing was implemented on perovskite samples over the last few years, demonstrating the impact of several parameters on the perovskite lifetime and stability. Even, if a lot of research work has taken place with indoor and outdoor tests on perovskite devices over the last years, the application of optoelectronic and spectroscopic techniques on perovskite samples at different degradation stages is still required for perovskite mini-modules tested outdoors. In this work, advanced indoor characterization of perovskite mini-modules was implemented at different degradation stages. Several spectroscopic and optoelectronic methods such as spatially resolved Electroluminescence/Photoluminescence, Dark Lock-In Thermography, Ultrafast and Raman spectroscopies have been employed for this purpose. The evolution of hotspots, shunt resistance and defects have been detected after outdoor degradation of perovskite devices. Moreover, changes in carrier relaxation mechanisms and in structural and chemical changes were studied on samples at different stages of degradation.

Keywords

Perovskite, Degradation, Electroluminescence, Photoluminescence, Dark Lock-In Thermography, Ultrafast Spectroscopy, Raman Spectroscopy, Outdoor Testing

Acknowledgments

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Ion Migration in Perovskite Solar Cells with Simulation and Experimental Analyses

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Abstract

Ion migration has challenged the practical applications of perovskite solar cells with undesirable hysteresis and degradation effect. We observe that the migration of cations from the perovskite layer towards the contact layer (CIS) is the main cause of high hysteresis and reduced efficiency. To better understand the behavior of *J-V* hysteresis under different scan rates and pre-bias conditions, the ionic patterns, recombination rates, and the electric potential were analyzed. The electron lifetime determined through V_{OC} decay experiments and simulations, and found that selecting appropriate scan rates is crucial for achieving high-efficiency and low-hysteresis perovskite solar cells. Additionally, it is found that device pre-biasing at a voltage close to its open circuit voltage can reduce cation migration from the perovskite/CIS interface towards the CIS contact, leading to higher efficiency and lower hysteresis. Conversely, Negative pre-bias voltages, can increase hysteresis and show inverted hysteresis behavior (-1 to -2 V) due to facilitating cation migration at the CIS side. Lastly, it is found from V_{OC} and simulation results that pre-conditioning within the range of milliseconds to seconds is important to have a highly efficient and hysteresis-free solar cell device.

Keywords

Perovskite Solar Cell, Ion-migration, Hysteresis, Pre-conditioning

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Improved Hole Extraction and Band Alignment *via* Interface Modification in HTM-free Ag/Bi Double Perovskite Solar Cells

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Abstract

In the relentless pursuit of sustainable energy solutions to power our ever-expanding array of smart devices, indoor photovoltaics (IPVs) have emerged as indispensable assets. Among the plethora of materials vying for attention, $Cs_2AgBiBr_6$, the double perovskite (DP), shines brightly for its exceptional characteristics: ease of processing, minimal toxicity, and remarkable stability, all tailored to meet the rigorous demands of IPV applications. In this poster, we present a hole transport material (HTM)-free $Cs_2AgBiBr_6$-based solar cells, wherein the surface of the DP is subjected to a n-butylammonium post-treatment, fostering a 2D/3D mixed interface. Concurrently, the conventional metal electrode and HTM components are replaced by a carbon black back electrode (CBE) derived from upcycled biowaste. This transformative modification of the 2D/3D interface not only mitigates charge recombination but also enhances band alignment at the perovskite/CBE interface. Furthermore, density functional theory (DFT) calculations elucidate that increasing the thickness of the 2D modification augments the likelihood of hole localization near the perovskite/CBE

interface, thereby facilitating their efficient extraction. Consequently, we obtain HTM-free solar cells with elevated power conversion efficiency, underscoring the efficacy of our low-cost, end-of-waste fabrication strategy.

Keywords

Silver-bismuth Double Perovskites, HTM-free Perovskite Solar Cells, Lead-free Perovskite Solar Cells,
Carbon-based Perovskite Solar Cells, Interfacial Engineering

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Improving the Performance and Stability of Triple Cation Perovskite Solar Cells Using Various 2D Passivation

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Abstract

Perovskite Solar Cells (PSCs) offer several advantages such as high carrier mobility, absorption coefficient, carrier lifetime, and low exciton binding energy. Over the past decade, their power conversion efficiency (PCE) has increased from 3.8% to an impressive certified value of 26.1%. However, the commercialization of PSCs faces challenges such as degradation from exposure to oxygen, moisture, and ion migration. To address these challenges, we employed a pin architecture, and various 2D passivation molecules between the interlayer of triple cation perovskite and the C_{60} were introduced. This was followed by sequential treatment of ammonium benzenesulfonate (ABS), Ethane-1, 2-diammonium iodide ($EDAI_2$), and phenethyl ammonium iodide (PEAI) to enhance the performance and stability of triple cation PSCs. The diammonium molecules act as a barrier to minority carriers and reduce contact-induced interface recombination through field-effect passivation. The ABS treatment is believed to have created an additional (PbI_2) on the film surface of triple cation perovskite $Cs_{0.05}(MA_{0.02}FA_{0.98})_{0.95}Pb(I_{0.98}Br_{0.02})_3$, which further reacted with PEAI to generate 2-dimensional perovskite layers. We observed that the sequential treatment of ABS, PEAI, and ($EDAI_2$) resulted in a significantly enhanced PCE of 24.03%, which is attributed to an increased open circuit voltage (V_{OC}) as a result of reduced non-radiative recombination.

Keywords

Perovskite Solar Cells, Power Conversion Efficiency, Triple Cation Perovskite, 2D Passivation,
Non-Radiative Recombination

Sequentially Hybrid Vacuum-Processed Multi-Cation Halide Perovskite

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Abstract

Multi-cation halide perovskites are very promising absorber materials for highly efficient solar cells. Thermal evaporation of perovskite films offers several advantages over solution-based deposition methods such as conformal growth and good control over film thickness and uniformity, also reflected in the widespread use of thermal evaporation in the semiconductor industry. However, we report on an interesting hybrid and a fully vacuum deposition method. The hybrid sequential deposition method consists of a solution-based deposition of a lead-containing precursor followed by vacuum deposition of formamidinium (FAI) to get a $FACsPbI_3$-perovskite film after thermal annealing. The Cs-component was either directly included in the precursor solution or sequential evaporated before the FAI evaporation with distinct differences in film formation. The fully vacuum process consists of three vacuum deposition steps followed by a thermal annealing step. The processes were recently reported to result in very efficient and robust perovskite layers and solar cells. In the X-ray diffraction data, one can see a reduced PbI_2 reflex at 12.7 °after the deposition of Cs and FAI and subsequent annealing. Moreover, the $FAPbI_3$-(100) reflex appears at 14.0 °so it can be concluded that the crystallization to the cubic α-phase perovskite has taken place. Details of process variations and their impact on film and device properties of solar cells in inverted p-i-n architecture will be discussed.

Keywords

Thermal Evaporation, Sequential, Hybrid, Multi-cation, p-i-n Architecture

Water-free PEDOT: PSS Formulation for Pb-Sn Mixed Perovskite Single-Junction and All-Perovskite Tandem Solar Cells

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Abstract

Single-junction Pb-Sn perovskite solar cells (PSCs) with a bandgap of 1.22eV are peaking at a power conversion efficiency (PCE) of over 23%. These efficiency values have been reached by following strategies to reduce non-radiative recombination losses by improving the quality of the absorber via additive incorporation and the interface with the electron transport layer (ETL) via surface passivation. Regardless, PEDOT:PSS is the standard hole transport layer (HTL) due to its high conductivity and selectivity. Its incorporation introduces drawbacks to the processing of perovskite devices, as it is typically processed from water-based dispersions, not being compatible with inert glovebox conditions. Additionally, it can permeate through the recombination layer and degrade the wide bandgap perovskite absorber (WBG), reducing reproducibility and increasing batch-to-batch variations in tandem cells. Due to the presence of PSS, the formed layer is acidic and hygroscopic, adding to the unfavorable stability of narrow bandgap perovskites and thus all-perovskite tandems. To tackle these problems, an anisole-based PEDOT:PSS formulation compatible with processing in the glovebox was introduced. By doing so, the WBG sub-cell never encounters ambient conditions. From contact angle measurements it is shown that the new PEDOT:PSS is more hydrophobic which is in agreement with the lower PSS content. Thickness measurements revealed an HTL thickness of 100 nm which can be tuned down by changing the processing parameters or diluting the coating solution. While thinner layers resulted in higher visual transmission, the increase in photocurrent generation was found to be negligible. Optical simulations are performed to analyze this behavior. At the same time, photoluminescence quantum yield (PLQY) measurements were conducted, showing a 60 mV increase in the implied open circuit voltage (iV_{OC}) in comparison to the standard device. The optimally thick HTL also positively impacts charge extraction, as can be seen from transient PL measurements. Finally, from current-voltage measurements an increased efficiency (from 19.4% to 23.3%) as well as improved reproducibility and shelf-life stability in all-perovskite tandem devices utilizing the new PEDOT:PSS formulation was observed. Tracking of the maximum power point revealed ≥ 21% stabilized efficiency, in contrast to a decreasing PCE for the standard device. Overall, our research provides insights into

the application of a water-free PEDOT:PSS dispersion which can substitute the standard PEDOT:PSS without compromising any of its benefits, but instead paving the way for more efficient, reproducible, and stable PSCs.

Keywords

Narrow Bandgap, All-Perovskite Tandems, PEDOT:PSS, Stability

Post-Lamination Treatment of Solvent-free Carbon Back-Electrodes for Fabrication of Efficient Perovskite Solar Cells

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Abstract

While substituting carbon for metals as the electrode material in perovskite solar cells (PSCs) enhances stability, reduces CO_2 footprint, and lowers production costs, carbon-based PSCs (C-PSCs) have struggled to surpass an efficiency record of 21% as of February 2024, trailing behind the overall PSC efficiency of 26%. The primary limiting factor often lies in the solvents present in the carbon paste commonly used for deposition, which can wash out the underlying charge transport layer (CTL), such as Spiro-OMeTAD in the *n*-*i*-*p* structure, thereby constraining CTL options. Another factor contributing to lower efficiency is the lack of back-reflection of escaped photons from the carbon layer. In addressing the first issue, we explored the press-transfer method for applying dried carbon films as an alternative to carbon paste to mitigate solvent-related damage. Laminated carbon films (CLam) were employed in an *n*-*i*-*p* architecture, revealing an excessively rough surface and uneven thickness. This resulted in a suboptimal connection between the carbon layer and the underlying surface during plate-to-plate pressing, leading to higher series resistance compared to blade coating or printing. To investigate this phenomenon, we utilized 3D laser scanning microscopy, mathematical modeling, and electro-optical characterizations. Through process optimization, we achieved an 83% efficiency compared to gold-based cells on a triple cation perovskite with a bandgap of 1.53 eV. However, even after optimization, concerns persisted regarding the roughness of the carbon layer potentially limiting device performance. To enhance electrode adhesion and interconnection quality, we employed solvents as post-lamination treatment. Employing the solvent treatment approach significantly reduced sheet and interface resistances, resulting in an efficiency exceeding 90% of gold-based reference cells. The reduction of sheet and interface resistances can be seen from *I*-*V* measurement and DLIT results.

Keywords

Carbon-based Perovskite Solar Cell, CPSC, Carbon Lamination, Solvent-free Carbon, Interface Resistance

Opportunities for FTO/NSG TEC™ Glass as a Functional Substrate in 3rd Generation PV

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Abstract

This research project investigates fluorine-doped tin oxide (FTO) and NSG TEC™ glass as a substrate for perovskite solar cells (PSC's). At present, tin-doped indium oxide (ITO) coated glass is one of the main substrates for laboratory prepared high performing solar cells. However, FTO is already mass-manufactured inexpensively via a chemical vapor deposition (CVD) technique and is the transparent conductive coating of choice for many industrial thin films. The glass conglomerate NSG commercially produces a range of TEC™ glass originally designed for solar control applications. Yet, it has shown excellent potential for the use as a substrate in PSC's. The CVD fabrication method avoids inconsistencies and difficulties related to the scaling up of lab-scale solution-based deposition methods such as spin coating or spray pyrolysis. The project will explore several different routes into investigating the substrate, including in-depth analysis and characterization with comparison against laboratory-built devices and, in particular, understanding the advantages over ITO. Modifications will be implemented to improve overall device performance, such as selective patterning of oxide layers to elevate charge transfer between an electrode and FTO at a series interconnection. Evaluation will be carried out over a range of conditions, with a particular focus on optimizing the characteristics of the blocking layer to enhance the performance of perovskite solar cells (PSCs).

Keywords

Fluorine-Doped Tin Oxide, Perovskite, Tin-Doped Indium Oxide, Chemical Vapor Deposition

Impact of Film Thickness on the Structural and Linear/Nonlinear Optical Properties in Highly Oriented $Cs_3Bi_2I_9$ Perovskite Films

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Abstract

This study presents the successful preparation of highly oriented $Cs_3Bi_2I_9$ films with different thicknesses using sol–gel spin-coating method. X-ray diffraction analysis confirmed a single hexagonal crystal phase with a high orientation along the direction. The crystallite size in the range of 27 and 41 nm increases with increasing film thickness. The bandgap energy ranged from 1.91 to 1.97 eV and was found to decrease with increasing film thickness, which was associated with the film relaxation with increasing film thickness. The refractive index, absorption index, complex dielectric coefficients, complex optical conductivity coefficients, and the ratio of free charge concentration also were found to increase with film thickness. The first-order linear optical susceptibility ranged from 0.22 to 0.38, the non-linear optical susceptibility $\chi^{(3)}$ and the nonlinear refractive index parameters $n^{(2)}$ were respectively within the range of 0.4–3.77 $\times 10^{-12}$ esu 0.96–7.97 $\times 10^{-11}$ esu, the highest value of all the parameters was found in the visible region at 500 nm. These findings position $Cs_3Bi_2I_9$ as a highly promising material for optoelectronics.

Keywords

Inorganic Lead-free Perovskite, Non-linear Optical Parameters, Highly Oriented Films, Dispersion Oscillator Analysis

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Passivating Inorganic Interlayers at the Perovskite/C_{60} Interface in Monolithic Perovskite Silicon Tandem Solar Cells

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Abstract

Metal halide perovskites have emerged in recent years as promising absorber materials for solar cells with the potential to combine high power conversion efficiency with low production costs. However, significant non-radiative charge carrier recombination occurs at the perovskite interface with the electron contact, the fullerene C_{60}, which prevents the full exploitation of the solar cell's potential. Thermal evaporation of the C_{60} contact layer induces states within the bandgap, which act as recombination centers, lowers the quasi-Fermi level splitting, and thus, limits the open circuit voltage (V_{OC}) in solar cell devices. Ultra-thin passivation layers at the perovskite/C_{60} interface are used to reduce non-radiative recombination losses. To enable industrial upscaling, our focus is on inorganic passivation layers deposited via atomic layer deposition (ALD, *e.g.*, AlO_x). By adjusting the ALD parameters, an AlO_x interlayer has been developed that increases the iV_{OC} up to 50 mV and improves the V_{OC} for single-junction and tandem devices. To better understand the effects that play a role in this passivation, photoluminescence quantum yield (PLQY), angle-resolved X-ray photoelectron spectroscopy (ARXPS), and surface photovoltage (SPV) measurements were carried out. Since state-of-the-art perovskite solar cells using a LiF_x passivation layer suffer from severe device degradation over time, initial stability testing was carried out providing indications that a thin AlO_x passivation layer can slightly improve device stability and thus, can serve as a robust alternative to LiF_x.

Keywords

Tandem Solar Cells, Perovskite/C_{60} Interface, Passivation Layers, Atomic Layer Deposition

Eco-profile Analysis of Printable Perovskite Solar Cell with Carbon Counter Electrode

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Abstract

Solar cells are a promising renewable source of energy for the mitigation of global warming and the emission of greenhouse gases. Decades ago, the commercialization of silicon solar cells began, and they have become a significant player in renewable energy scenes. Life Cycle Assessment (LCA) studies done on silicon solar cells show that CO_2 emissions from their production process range from 14 to 73 g CO_2-eq/KW depending on the electricity mix used for the production. However, despite the emissions being less than those of fossil fuels they are still not negligible when considered to meet the global energy demand objectives. Perovskite solar cells are an efficient competitor for silicon solar cells, especially in terms of the carbon footprint, cost-effectiveness, and recyclability. Since the first realization of perovskite solar cells, many efforts have been taken to increase the PCE, stability, and durability. In our work, we develop a perovskite solar cell architecture with enhanced stability and competing PCE employing different HTL, such inorganic $CuInS_2$ and organic P3HT. Except for the HTL layer deposited by spin coating, the complete device stack was deposited by blade coating method out of the glovebox. To improve the eco-profile of the device, we used low-temperature printed carbon as a counter electrode instead of gold, which has a significant environmental impact. The cradle-to-gate LCA is performed based on detailed primary data of the lab scale process for the evaluation of the environmental performances of the device.

Keywords

Carbon Electrode, Life Cycle Assessment, Printable Perovskite Solar Cells, Cradle-to-Gate

Loss Analysis of a Perovskite/Perovskite/Silicon Solar Cell

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Abstract

Perovskite/perovskite/silicon triple junction solar cells are a rather new architecture that offer a potentially cost-effective and highly efficient solution. Additional junctions reduce thermalization losses, yielding a detailed balance limit of 49.6% for this cell architecture. Simulations aid in identifying factors contributing to efficiency loss, guiding developers to improve specific layers. Current key challenges include achieving stable perovskite compositions at the right bandgap, as well as finding suitable hole and electron extraction and recombination layers. In this work, we first analyze the optical efficiency of a perovskite/perovskite/silicon triple junction solar cell. Our simulation is validated on experimental data, which we regard a representative cell for the current cells of this architecture. For this triple junction cell design, we identify the most impactful loss by the current mismatch between the cells, which is a loss of approximately 4 mA/cm^2. By using perovskites with optimized bandgaps and/or adjustments of the perovskite layer thicknesses, this can be resolved. Reflection causes 10% of the incoming light to be lost. A textured front surface can mitigate this. Also, the interlayers absorb 8% of the incoming photocurrent parasitically, which can be lowered by using the thinnest layers possible. By implementing the mentioned improvements, we predict a short-circuit current of 14.1 mA/cm^2 and an open-circuit voltage of 3.48 V with an efficiency of 44.3%, assuming idealized electrical properties. To assess the electrical losses, the top and middle perovskites are investigated as individual single-junction cells. With an opto-electrical model, we can investigate the internal processes and parameters and analyze the impact on the power conversion efficiency (PCE) with band diagrams and *J*-*V* curves. These internal parameters are validated by several different experiments. We investigate carrier lifetimes, surface recombination velocities, band alignments, series resistances, and the impact of preconditioning.

Keywords

Simulation, Sentaurus, Perovskite, Silicon, Multijunction

Unraveling the Role of Traps in Understanding the Superlinear Power Law and Vacancy-Assisted Ion Conduction in Hybrid Organic-Inorganic Metal Halide Perovskite

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Abstract

In cutting-edge energy research, organic-inorganic metal halide perovskites (OIMHPs) are at the forefront. Therefore, understanding how standard operating conditions, such as air temperature, humidity, light exposure, *etc.*, affect the transport in OIMHP-based energy harvesting devices is crucial. Here, we have examined the temperature-dependent complex impedance, complex electric modulus, and AC conductivity spectra of an OIMHP, $FAPbBr_2I$. We have used theoretical models, such as the Havriliak Nigami model and the Jonscher Power law, to fit the obtained results. The Maxwell-Wagner equivalent circuit model has been utilized to decipher the resistive and capacitive contributions of grains and grain boundaries to the total impedance at various temperatures. The AC conductivity spectra exhibit distinct behaviors in two different temperature zones, viz. at low-temperature (LT) and high-temperature (HT) zones. Normal ionic conduction with sublinear dispersion was discovered in the LT region (323-403 K) where the frequency exponent (n) < 1. In the HT regime (413-463 K), the sublinear dispersion transforms into a superlinear dispersion (n) >1. The appearance of a superlinear (SPL) AC conductivity dispersion beyond a critical temperature of 413 K is ascribed to the decoupling or the release of a large number of charge carriers from the trap centers in the HT regime. Additionally, we notice an immediate increase in DC conductivity beyond 413 K, which can be attributed to the dominance of vacancy-assisted ionic conduction because, when the charge carriers are discharged, the vacancies constituting the trap states become accessible for ionic conduction. Moreover, by investigating the temperature-dependent space charge limited current-voltage characteristics of $FAPbBr_2I$ single crystal, we have unveiled the role of trap states in understanding the SPL behavior and vacancy-mediated ionic conduction. Finally, it is noted that although the complex electric modulus and AC conductivity scale to a master curve in the LT regime, the scaling leads to the non-overlapping curves in the dispersive high-frequency region of the HT regime, further supporting the existence of two distinct dispersion behaviors and conduction mechanisms in the low- and high-temperature regimes.

Keywords

Single Crystal, Superlinear Power Law, Ionic Conduction, Space Charge Limited Current, Dielectric Relaxation, Conductivity Scaling

Low-Temperature Hole-Transport Layers' Investigation for Inverted Flexible Perovskite Solar Cells

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Abstract

Halide perovskite (HP) photovoltaics (PV) demonstrate remarkable efficiency exceeding 26% for terrestrial applications. HP semiconductors demonstrate a unique combination of optical and transport properties in micro-crystalline thin films: diffusion length (>1 μm), lifetime (up to 1 ms), and direct-bandgap structure. The potential of perovskite solar cells lies in their ability to be fabricated on glass or plastic substrates, offering advantages such as lightweight, portability, and suitability for integration on curved surfaces. Although technologies for flexible solar cells based on silicon, cadmium-tellurium (CdTe), and copper-indium-gallium selenide (CIGS) exist, there is no well-developed and widespread approach for perovskite-based flexible solar cells (SC). One of the key challenges in designing flexible SC is the necessity for low-temperature conductive layers due to the limited thermal stability of plastics (< 150 ℃). This demands meticulous fabrication of charge transporting layers and absorber films to maintain high output performance. Particularly, modification of highly efficient hole transporting thin films based on Nickel oxide or application of organic conductive layers can be used. In this work we made a complex investigation for the variation of hole-transport layers with organic conductive material MeO-2PACz deposited by spin coating and explored NiO_x deposition using ion-beam sputtering – the oxidation of dispersed nickel particles, followed by post-treatment annealing at a temperature of 120 ℃. The impact of the HTL type on the output characteristics of flexible solar cells was estimated under the light of a solar simulator. The benefits of the used methods of growth flexible HP solar cells were discussed.

Keywords

Flexible Perovskite Solar Cells, Low-Temperature Manufacturing, Hole-Transport Materials, Ion-Beam Sputtering

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Fabrication of Hermetically Laser-Sealed Printable Perovskite Solar Devices Towards Superior Extrinsic Stability

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Abstract

A hermetic encapsulation is required to protect perovskite solar cells (PSCs) from the most relevant sources of degradation – humidity and oxygen. According to the IEC61646 PV standard test, commercial photovoltaic devices must be stable from - 40 °C to 85 °C and relative humidity of 85%. Therefore, to achieve the previously mentioned requirements, the PSCs should be fabricated with thermally stable layers and protected by a long-term stable hermetic encapsulation. Previously, we reported a laser-assisted glass frit encapsulation that successfully achieved long-term stability for PSCs with n-i-p and HTM-free structures. An advanced dual laser beam glass frit sealing process was previously developed and optimized to hermetically encapsulate n-i-p PSCs at 65 ± 5 °C for a short processing time of < 60 s. In contrast, printable HTM-Free perovskite solar cells have been reported to be sealed with a single laser beam at 100 °C for a long processing time of *ca.* 35 min. Therefore, this work aims to use the dual laser sealing process to hermetically encapsulate HTM-Free PSCs and mini-modules. Moreover, a 100 x 100 cm^2 panel (*ca.* 234 cm^2) was manufactured using dual laser-sealed mini-modules (*ca.* 6 cm^2). The novel sealing process had no impact on the performance of the encapsulated devices, since the power conversion efficiency (PCE) of both small-area PSCs and mini-modules slightly increased from (8.27 ± 0.83)% to (10.75 ± 1.41)% and from (6.08 ± 0.32)% to (6.61 ± 0.49)%, respectively. After sealing 39 mini-modules in series and in parallel, the assembled panel delivered an average PCE of 4.71% when exposed to 789 W m^{-2} solar irradiation. In conclusion, this study indicates that dual laser sealing has a low impact on the performance of lab-scale devices and mini-modules, and it also reinforces that this sealing procedure can be suitable for encapsulating large-area PSCs.

Keywords

Perovskite Solar Cells, Hermetically Laser-Assisted Glass Frit Encapsulation, Stability, Scalability

Acknowledgments

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Transient Electroluminescence in Perovskite Devices: The Role of Ion Migration

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Abstract

Perovskite solar cells (PSCs) show great promise to compete in the photovoltaic market, thanks to their outstanding performance as the light absorber material in thin film solar cell stacks, their simple solution-processing and low-cost fabrication process. Similarly, perovskite light-emitting diodes (PeLEDs) are a promising technology for efficient and cost-effective lighting and displays. To further optimize the performance, stability, and reliability of perovskite devices, detailed characterization and understanding of performance losses over time are required. This includes examining the recombination pathways of charge carriers and how these change with the migration of mobile ions, which can be a major limiting factor for achieving (and maintaining) the highest possible performance. Metal halide perovskite semiconductors show mixed ionic-electronic conductivity. Mobile ions can slowly redistribute under light or voltage, in timescales ranging from milliseconds up to minutes or hours. This study focuses on the impact of mobile ion redistribution on charge recombination processes, by looking at the transient electroluminescence (TrEL) profiles of perovskite devices. The inclusion of mobile ions in drift-diffusion simulations is found to be crucial to reproducing the experimental TrEL responses of PSCs and PeLEDs. These findings contribute to the understanding of transient ionic processes in perovskite-based devices.

Keywords

Perovskite, Solar Cells, Light Emitting Diodes, Pulsed Operation, Simulation, Transient Electroluminescence

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Understanding Crystal Growth Dynamics of Perovskite on Textured Silicon Substrates for Multijunction Solar Cell Applications

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Abstract

In the industry, double-sided textured (micro-meter sized pyramid) silicon is commonly produced to decrease reflection losses and improve light trapping. Solution-based processing methods of perovskite on top of the textured Si showed low conformality, which resulted in shunts and non-working solar cells. To overcome this issue, physical vapor deposition (PVD) is used for the different precursors to conformally coat the perovskite on the silicon pyramids. In this study, we investigate a processing method namely the hybrid route, in which inorganic precursors are co-evaporated using the thermal vapor deposition technique and then organic precursors including additives are spin-coated, followed by a thermal annealing. By doing that, high conformality of high-quality perovskite thin films on top of the textured silicon is achieved as well. In the hybrid route, we also study the crystallization dynamics at different stages of the hybrid route and the effect of grain sizes on FF and open circuit voltage (V_{OC}).

Keywords

Hybrid Route, Additives, Perovskite Silicon Tandem Solar Cells, Crystallization

Electrophoretic Deposition of Potassium Sodium Niobate Thick Perovskite Coatings for Energy Harvesting Applications

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Abstract

Globally, depleting non-renewable energy resources and environmental pollution are significant challenges. Researchers and scientists have recently turned their focus to perovskite coatings because they have the potential to replace green fuel production with maximum efficiency without environmental hazards. KNN ceramics are fabricated by solid-state method, calcined at 850 ℃, and electrophoreticcally deposited on Ni-substrate uniquely then, sintered at high temperatures. The X-ray diffraction and FTIR confirmed the development of a pure KNN perovskite structure and metallic bond groups (-O-Nb-O) present in the coating, respectively. The sintering temperature resulted in the pronounced peaks observed in KNN ceramics, confirmed by Raman spectroscopy, and easily observed in SEM having "square" and "circular" morphology with grain growth. The coating thickness was measured around 120 μm with increasing deposition rate (0.40 μm/sec) was calculated. The coating roughness (~813 nm) was confirmed by atomic force microscopy. Complex impedance spectroscopic (CIS) analysis confirmed the high dielectric constant (~4789) with a high transformation and curie temperature ($T_{O\rightarrow T}$ ~ 280 ℃ & T_C ~ 480 ℃), respectively. The increasing conductivity (≥830 μS/m) at higher frequencies and temperatures agrees with the hopping conduction mechanism, which confirmed the negative temperature coefficient of resistance (NTCR). This work holds great significance in photovoltaics, sensors, actuators, spintronics, and energy harvesting applications.

Keywords

Perovskite, Electrophoreticcally, Dielectric, Sensors, Actuators, Spintronics

Acknowledgments

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Efficient Role of Brominated Metalloporphyrin Additive in Improvement of Performance and Stability of Carbon-Based Planar Perovskite Solar Cells

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Abstract

Despite the great promotion of perovskite solar cells, the polycrystalline nature of the perovskite layer leads to the formation of defect trap states at surfaces and grain boundaries, hampering photovoltaic functionality. Motivated by porphyrin molecules tunable optoelectronic characteristics, we introduce zinc(II)*β*-tetra-bromo-meso-tetra-phenyl-porphyrin as an additive in the perovskite layer to improve the performance and stability of perovskite solar cells. The electronegative bromine atoms alter the distribution of electron density within the metalloporphyrin. Through Fourier transform infrared spectroscopy (FTIR) analysis, it has been observed that there is an interaction between the modified electron density of metalloporphyrin and the charge trap state, particularly the under-coordinated Pb^{2+} deep traps. The potential for passivating defect states has been confirmed through space-charge limited current (SCLC) measurements using electron-only devices and a quantitative assessment of trap density. Moreover, the V_{OC} alteration in response to light intensity suggests a reduced rate of trap-assisted recombination and effective prevention of electron trapping in the vacancy sites of the crystal structure in the modified layer. Additionally, XRD analysis demonstrated a notable increase in the intensity of peaks associated with the perovskite crystal structure within the modified perovskite. On the other hand, besides the positive impact on the quality of perovskite crystallinity, validated by the UV-Vis absorption and photoluminescence spectrum, the energy levels alignment between the perovskite, metalloporphyrin, and hole transport material leads to the development of a graded band structure to assists the efficient transport of electron-holes towards the hole transport layer, as evidenced by the photoluminescence spectra and the SCLC measurements. As a result, the efficiency of the best device is up to 18.5%, a factor of 15% increase to that of the reference cell with a value of 16.1%, which is superior in planar device structure with copper indium disulfide (CIS) as a hole transport material and carbon as back contact. Furthermore, enhanced hydrophobicity and crystalline quality improve the stability of devices, and the modified device maintained 96% of its initial efficiency after 40 days in comparison with the control device, which displayed a 38% drop in its performance.

Keywords

Planar Perovskite Solar Cells, Metalloporphyrin, Additive, Grain Boundary, Passivation, Defect State, Charge Transfer

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Design of Photovoltaic-grade Interface Passivation Layers to Enhance the Efficiency of the Perovskite Solar Cells

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Abstract

The efficiency of Perovskite solar cells (PSCs) has shown remarkable progress in recent years, making them promising candidates for next-generation photovoltaic technology. However, challenges remain in achieving stable and high-efficiency performance, particularly concerning interface passivation. This study focuses on the design and optimization of photovoltaic-grade interface passivation layers to enhance the efficiency of PSCs. A comprehensive investigation into the effects of different passivation materials, deposition techniques, and interface engineering strategies is conducted to elucidate their impact on device performance. The role of using the NH_4Cl and PMMA: PCBM passivation layers in mitigating charge recombination processes at the perovskite/electron transport layer and perovskite/hole transport layer interfaces is elucidated. Additionally, the influence of passivation on device stability and hysteresis effects is investigated to ensure long-term performance reliability. The results demonstrate that tailored passivation layers effectively suppress interface recombination, leading to remarkable long-term stability after 116 days (2784 hours) of storage under dark conditions and in Ar ambient, achieving a PCE of 19.4%. Moreover, insights gained from this study contribute to a deeper understanding of the underlying mechanisms governing PSC operation and provide guidelines for the rational design of interface passivation strategies. Ultimately, this research facilitates the development of high-performance PSCs with enhanced efficiency, stability, and commercial viability, thereby advancing the prospects of perovskite-based photovoltaics as a sustainable energy solution.

Keywords

Passivation, Electron Transport Layer, Long-Term Stability

Acknowledgments

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Investigations on Additives for Batch Cluster Processing for Industrial TOPCon Solar Cells

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Abstract

Industrial Perovskite-Silicon tandem solar cells require a cheap and efficient Si bottom cell. This paper focuses on the Si bottom cell. Tunnel Oxide Passivated Contacts (TOPCon) solar cells are the next industrially relevant cell concept after the Passivated Emitter Rear Contact (PERC) concept. TOPCon cells require a poly-Si film on the backside, which is also deposited on the sides and on the front of the cell. The parasitic deposition at the edges and front side needs to be removed in order to guarantee a safe module performance. This is done using wet-chemical etching steps. First, the phosphorous-doped Si glass (PSG) on the poly-Si, formed during the exSitu doping of poly-Si, is removed from the front and the sides of the wafer in a single side inline HF wet process step. Next, the exposed poly-Si film is removed in a KOH batch process. Here, the 15 to 30 nm thin PSG layer on the rear side acts as a masking layer to protect the poly-Si layer underneath. This is only possible by using an additive, which enhances the Si/SiO_2 etch rate selectivity in KOH in order to maintain the PSG acting as a masking layer. This additive is the main driving parameter of the running cost for chemical edge isolation and poly wrap around removal step and hence an important factor for the CoO for TOPCon cells manufacturing. This work focuses on such additives. A newly developed additive is compared to a market additive in terms of Si/SiO_2 etch rate selectivity, Si etch rates, and Si surface morphology after polishing. It is shown that etch rate selectivity and Si polishing grade are sufficient for both additives, whereas the newly developed additive shows a 20% increased Si etch rate. This is beneficial because the increased Si etch rate allows lower process time and KOH usage, resulting in lower costs, higher throughput, and less water consumption.

Keywords

Silicon Solar Cell, TOPcon Cluster, Additive, PSG, Wet Processing, Industrial, Si Bottom Cell

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Physical Vapor Deposition of Tin- and Lead-Based Halide Perovskites Via *in situ* X-ray Diffraction: From Phase Evolution to Formation Kinetics to Thin Film Solar Cells

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Abstract

This study explores advancements in organic-inorganic lead-free perovskite materials, focusing on the film formation process during deposition by thermal evaporation for lead- and tin-based perovskites. It involves growth analysis, solar cell component preparation, and the analysis of optoelectronic properties. Key aspects include *in situ* X-ray diffraction for observing phase changes during growth, analysis of perovskite degradation, and development of multi-stage all-vacuum growth processes for multi-component perovskite thin films. State-of-the-art Pb-based perovskites and potential Sn-based compounds will be examined. We conduct sequential deposition/reaction processes under controlled temperature and flux variations in order to estimate reaction constants, diffusion constants, sticking coefficients, and activation energies. *In situ* X-ray diffraction in real-time will be the primary experimental method, providing insights into phase formation during thin film growth. A sandwich-like material structure design (*e.g.*, PbI_2-CsI-SnI_2) will be used to analyze the perovskite's growth mechanism and diffusion model as well. Further exploration involves understanding how IA-group cations like Cs achieve thermodynamic and kinetic balance in PbI_2 and SnI_2, facilitating the formation of stable and high-quality perovskite layers. There are three possible film growth mechanisms: (a) Precursor vapor reaction: Solid-state precursor films react from the vapor phase to form the perovskite film. (b) Diffusion couple reaction: Two or more solid-state precursor films react via chemical diffusion. (c) Exchange reaction: Another component fully or partly exchanges one component (anion or cation) during annealing processes (*e.g.*, $MAPbI_3$ to $MAPbBr_3$ exchange reaction). This study aims for solvent-free, controllable film deposition, utilizing sequential evaporation for better reproducibility. Different film growth mechanisms will be investigated, including precursor vapor reaction, diffusion couple reaction, and exchange reaction. Both Pb-based perovskites and Sn-based compounds are investigated, with a focus on reaction kinetics and diffusion. The ultimate goal is to advance toward the production of large-area, high-efficiency solar cells through physical processes, including the completion of various layers within a vacuum chamber, paving the way for stable and high-performance commercial applications in solar energy technology.

Keywords

Halide Perovskites, Physical Vapor Deposition, *In Situ* X-Ray Diffraction, Co-evaporation, Diffusion Model, Precursor Vapor Reaction, Exchange Reaction, Perovskite Solar Cells

All-Inorganic $CsPbBr_3$ Perovskite Solar Cells Via Sequential Thermal Evaporation

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Abstract

Perovskite solar cells (PSCs), advancing solar technology with remarkable photoconversion efficiency (PCE) and stability, typically use hybrid organic-inorganic lead halide perovskites. However, concerns remain about the organic component's impact on degradation. Transitioning to all-inorganic cesium perovskites is an alternative route to tackle the long-term stability challenges in PSCs. Within inorganic perovskites, $CsPbI_3$ suffers from polymorphism ranging from the photoactive α-phase to the inactive δ-phase. In contrast, $CsPbBr_3$ perovskites offer robust thermal, humidity, light stability and do not suffer from polymorphism. With a Shockley-Queisser single-junction limit of ~ 16% and a wide bandgap of 2.3eV, it is attractive for semi-transparent, building-integrated photovoltaics, and multi-junction applications. Many $CsPbBr_3$ works are based on solution-processing using conventional spin coating technique, limiting uniformity over large areas. Also, dissolving the precursors in solution, which frequently comes with toxicity concerns, can be challenging. Alternatively, thermal evaporation offers a solvent-free, industry-compatible fabrication method, enabling precise thickness control, conformal and uniform coverage over large substrates. Here, we fabricate a solvent-free $CsPbBr_3$ PSC via dual-source sequential evaporation. $CsPbBr_3$ films, deposited on compact SnO_2 electron transport layer, are pinhole-free and exhibit phase purity with reduced defects. Thin film annealing studies using X-ray diffraction, conducted alongside device investigations, revealed a decrease in phase transition temperature from 300 ℃ to 250 ℃. Finally, the fabricated device results in a PCE of 5.6% with an open-circuit voltage of 1.4V. An all-inorganic PSC with a vacuum-processed absorber layer is demonstrated to achieve a phase-pure, compact film of the desired thickness, paving the way for exploring $CsPbBr_3$ active layer.

Keywords

Evaporated Perovskite Solar Cells, All-inorganic Perovskite Solar Cells, Wide Bandgap Photovoltaics, Semi-transparent and Building-integrated PV

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Impact of Ion Migration on the Performance and Stability of Perovskite-Based Tandem Solar Cells

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Abstract

The stability of perovskite-based tandem solar cells (TSCs) is the last major scientific/technical challenge to be overcome before commercialization. Understanding the impact of mobile ions on the TSC performance is key to minimizing degradation. Here, we present a comprehensive study that combines an experimental analysis of ionic losses in Si/perovskite and all-perovskite TSCs using scan-rate-dependent current-voltage (J-V) measurements with drift-diffusion simulations. Our findings demonstrate that mobile ions have a significant influence on the tandem cell performance, lowering the ion-freeze power conversion efficiency from >31% for Si/perovskite and >30% for all-perovskite tandems to around 28% in steady-state. Moreover, the ions cause a substantial hysteresis in Si/perovskite and all-perovskite TSCs at high scan speeds (400 V/s and 40 V/s, respectively), and significantly influence the performance degradation due to field screening. Additionally, for all-perovskite tandems, subcell-dominated measurements reveal more pronounced ionic losses in the wide-bandgap subcell during aging, which we attribute to its tendency for halide segregation. This work provides valuable insights into ionic losses in perovskite-based TSCs, which helps to separate ion migration-related degradation modes from other degradation mechanisms and guides targeted interventions for enhanced subcell efficiency and stability.

Keywords

Perovskite-based Tandem Solar Cells, Mobile Ions, Ionic Losses, Wide-bandgap, Low-bandgap, Subcell Dominating Fast Hysteresis Measurements, Light Aging, Degradation

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Machine Learning to Analyze the Accelerated Aging of Perovskite Solar Cells

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Abstract

Perovskite solar cells have improved in efficiency and stability over the last decade, but we still lack commercially viable perovskite solar cells. To estimate the carbon footprint and energy returns, it is crucial to understand the lifetime of solar cells or the total energy yield. In this study, we are planning to do accelerated aging to understand the breaking points of perovskite solar cells. With the help of machine learning and drift-diffusion models, we are trying to extract the parameters of solar cells from the J-V plot. A machine learning model will analyze a series of J-V curves and give the insights that lead to change in J-V. The machine learning models were trained using the data generated by drift-diffusion model. While the applicability of this approach to experimental data is still being investigated, successfully generalizing the model to experimental data could enable us to analyze the causes of degradation. This, in turn, could be enhanced through interface engineering and material optimization.

Keywords

Stability, Accelerated Aging, Machine Learning

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Study of Reverse Bias Degradation in Perovskite Solar Cells and Modules

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Abstract

Perovskite solar cells have witnessed remarkable efficiency gains over the last decade, now matching crystalline silicon at 26.1%. However, their scalability is hindered by stability concerns, notably their susceptibility to prolonged stress, particularly reverse bias voltages. Reverse bias or negative voltages can build up over cells interconnected in series due to a difference in photocurrent generation in the cells. The difference in photocurrent can be caused due to defects in the cells, or more commonly to a difference in illumination on the cells caused by partial shading. Shading can occur due to nearby buildings, trees, and other objects that can block a part of the incoming sunlight. In this study, the effects of reverse bias voltages in cells and modules interconnected in series are investigated. The cells in the module are interconnected in series with laser scribing technique using P1, P2, and P3 scribe. The aim is to understand the effects of the scribe on the reverse bias degradation of the modules. The scribing introduces new factors into the samples, such as defects near the scribing area, and it could be a starter point for degradation. Similar tests with other thin film technologies, like CIGS, reveal that degradation is initiated from the scribes, and our preliminary tests reveal a similar phenomenon with perovskites. Perovskites are known to have low reverse breakdown voltages, but the degradation can initiate at voltages lower than the breakdown voltage. Reverse bias stresses are applied on the modules mainly in two ways, altering scan speeds in a reverse scan and at constant reverse bias voltages. For further analysis, some visual characterization techniques, like electroluminescence and thermal imaging techniques, are utilized to understand the degradation phenomenon in the samples. Electroluminescence would provide insights about the deterioration of the cells, and hotspot creation can be studied with infra-red imaging.

Keywords

Perovskites, Reverse-bias, Reliability, Degradation, Shading, Modules, Up-scaling

Solution Processability of Cu_2AgBiI_6 Films for Flexible Photovoltaic Applications

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Abstract

Lead-based perovskites have shown remarkable potential for photovoltaic applications. However, due to the environmental concerns and toxicity issues related to lead, there is a growing interest in reducing the use and increasing recycling of hazardous chemicals, while exploring lead-free alternatives. In this study, we investigate a lead-free perovskite-inspired material as a promising photo-active layer for indoor photovoltaic applications. We focus on a lead-free perovskite, inspired material that is made from copper silver bismuth iodide (Cu_2AgBiI_6, CABI). Recent studies demonstrate that this material exhibits favorable properties for indoor photovoltaic applications. It possesses a suitable bandgap for efficient light absorption under indoor lighting conditions. In addition, CABI exhibits good stability under ambient conditions, making it an interesting candidate for indoor photovoltaic applications. This material has proved to be solution-processable, allowing the use of flexible substrates, low-cost, and scalable fabrication techniques. We employed facile solution processing routes such as spin coating, slot-die coating, and gravure printing to fabricate thin films of CABI. We utilized techniques such as UV-Vis spectroscopy and scanning electron microscope to analyze the material's light absorption characteristics and thin film morphology. Furthermore, device fabrication on flexible substrates (PET/ITO/SnO_2/CABI/P3HT/Au) proved to yield similar performances as those devices fabricated on glass. In conclusion, our study demonstrates the promise of CABI as a sustainable and eco-friendly alternative for lead-based perovskites. Preliminary results highlight the potential of CABI for use in indoor energy harvesting photovoltaic applications.

Keywords

Lead-Free, Perovskite-Inspired Materials, Solar Cells, Flexible Substrates

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Hybrid TiO_2-TiAcAc/SnO_2 Electron Transporting Layer Enable High V_{OC} in Carbon-based Perovskite Solar Cells

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Abstract

Carbon-based perovskite solar cells (C-PSCs) are attracting considerable attention in the field of photovoltaic technology due to their potential for cost-effective production and exceptional long-term stability. However, a key barrier to their growth is the limited power conversion efficiency (PCE). This study demonstrates an approach to improve the PCE of C-PSCs by introducing a hybrid TiO_2+TiAcAc/SnO_2 electron transporting layer through a low-temperature process (≤150°C). The technique involves incorporating titanium diisopropoxide bis(acetylacetonate) (TiAcAc) into TiO_2 and modifying the interface between the electron transporting layer (ETL) and perovskite by using an ultrathin SnO_2 film. The addition of TiAcAc effectively closes the voids among the TiO_2 nanoparticles, resulting in a more compact TiO_2 coating. Consequently, the inclusion of TiAcAc in TiO_2 leads to an augmentation of the open circuit voltage (V_{OC}) to 1.10V. Moreover, an ultrathin SnO_2 film is applied to passivate the interface between TiO_2+TiAcAc and the perovskite layers. This results in an increased fill factor (FF) by reducing interface defects. Therefore, the device with an active area of 0.09 cm^2, utilizing a TiO_2+TiAcAc/SnO_2 ETL and fabricated under ambient conditions with a relative humidity of 35%RH, achieves a maximum PCE of 16.00%, with an open-circuit voltage (V_{OC}) of 1.09V, a short-circuit current density (J_{SC}) of 20.34 mA cm^{-2}, and a fill factor (FF) of 72.00%. It is expected that this work will offer an effective approach to raise the photovoltaic performance of the C-PSCs.

Keywords

Carbon-based Perovskite Solar Cells, Electron Transporting Layer, Tio_2, TiAcAc, SnO_2

Sustainable Planar HTM-Free Carbon Electrode-Based Perovskite Solar Cells: Stability Beyond Two Years

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Abstract

Swift advancement in perovskite solar cell (PSCs) efficiency poses a challenge in maintaining a balance among sustainability, efficiency, and cost for competitive commercialization. Ongoing research is dedicated to effectively addressing these challenges. Traditional PSCs rely on expensive and unstable hole-transporting materials (HTMs) and noble metal electrodes, leading to poor device stability. To overcome these challenges, this study introduces unencapsulated planar HTM-free carbon electrode-based PSCs (C-PSCs) created through an entirely low-temperature process (< 160 ℃) in ambient atmospheric conditions. The approach emphasizes simplicity and cost-effectiveness, incorporating a single electron transporting layer and a one-step perovskite layer ($Cs_{0.17}FA_{0.83}Pb(I_{0.83}Br_{0.17})_3$) fabrication. Carbon films, prepared using an ethanol solvent interlacing method and heat-press transfer, serve as both hole transport layers (HTL) and electrodes. This simplified architecture leverages the properties of carbon materials, achieving the highest power conversion efficiency (PCE) of 11.09% and exceptional shelf-life stability exceeding 2 years (~20,000 hours) without encapsulation. Remarkably, thermal and humidity stability tests under accelerated aging conditions (85% relative humidity, 85 ℃) demonstrated an average 90% efficiency drop after 100 hours. Furthermore, the scalability of the technique is demonstrated in 1.00 cm^2 planar HTM-free C-PSCs on recycled FTO/TiO_2-NPs substrates, exhibiting remarkable performance under both 1 sun and LED illuminations. This approach lowers production costs, making PSCs more renewable and sustainable, paving the way for cost-effective and eco-friendly commercialized PSCs.

Keywords

Carbon Electrode, HTM-free, Low-temperature, Perovskite Solar Cells, Stability, Sustainability

Strong Impact of Substituent Position in PEAI-Founded Organic Cations to Enable the Efficient and Durable 3D/2D-Constructed Perovskite Solar Cells

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Abstract

The passivation of perovskite solar cells (PSCs) is inevitable to improve their performance and stability. Integrating 2D-forming phenylethylammonium iodide (PEAI) salts for passivation is an emerging strategy due to their hydrophobic character and improved stability in PSCs, although various cations have been implemented. This study investigates the impact of 9 different large organic cations, particularly halogenated with fluorine (F), chlorine (Cl), and bromine (Br) as the substituents at -*ortho* (-*o*), -*meta* (-*m*), and -*para* (-*p*) positioned-PEAI salts (coded as *x*-X-PEAI where *x: o*, *m*, *p* and X:

F, Cl, Br) synthesized using a straightforward method on the passivation of 3D perovskite surfaces and their subsequent effects on device performance and stability. The formation of 2D layers on top of the 3D perovskite was confirmed using X-ray diffraction (XRD) and grazing-incidence wide-angle X-ray scattering (GIWAXS) analyses for all cations, regardless of the nature and position of the halogen. Density functional theory (DFT) analysis was employed to understand the underlying mechanisms behind the observed performance differences. It revealed that m-substituted cations exhibited lower formation energies and higher interfacial dipoles, leading to enhanced device performance compared to their -*ortho* and -*para* counterparts. Among the halogenated PEA+ iodide salts tested, the device treated with *m*-BrPEAI exhibited the highest efficiency of 23.42%, with a high open-circuit voltage (V_{OC}) of 1.13 V and fill factor (FF) of 81.2%. However, considering overall efficiency, stability, and reproducibility, the treatment with *m*-ClPEAI salt yielded the best performance. This comprehensive study contributes to understanding surface passivation in PSCs and offers insights for optimizing device performance through the rational design of large organic cations.

Keywords

3D/2D Perovskite Solar Cells, Substituent and Position Effects, Stability, Large Organic Cations

Analysing Instability Mechanisms of Perovskite Solar Cells with 2D/3D Interfaces Under Light- and Heat-Operational Condition

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Abstract

Two-dimensional/three-dimensional interfaces (2D/3D) are widely used in perovskite solar cells to obtain excellent optoelectronic properties. It improves long-term stability under mild conditions but becomes destructive in harsh aging conditions. Here, we show that the Ruddlesden-Popper (RP) perovskite is intrinsically unstable and gradually decomposes into PbI2 and metallic lead (Pb^0) under photo-thermal aging. The RP/3D structure collapse leads to a rise in vacancies that force iodide ions to migrate readily to the anode. The anode undergoes a redox reaction with iodide/polyiodide, leading to a decrease in mobility and doping concentration of the transport layer and a sharp increase in the series resistance. Dion-Jacobson (DJ)/3D is found to be much more structurally stable and effective in blocking the iodide migration process. However, its rebound of iodide causes iodide to form an interstitial defect in the bulk, exacerbating non-radiative recombination. Finally, introducing DJ to the 3D grain boundary rather than the top surface can confine the mobile ion inside the grain and suppress the cation phase segregation. As a result, the ultra-stable perovskite solar cells have an extrapolated T_{80} of over 1000 hours at 85 ℃ and 1 sun.

Keywords

Perovskite Solar Cells, Two-Dimensional, Operational Stability, Interfaces, High Temperature, Light-Soaking