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OPERATIONAL STABILITY OF INVERTED PEROVSKITE SOLAR CELLS UNDER INERT AND VACUUM CONDITIONS: THE ROLE OF ION MIGRATION, ELECTRODES

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ABSTRACT

The long-term operational stability of perovskite solar cells (PSCs) remains a critical challenge for their commercialization, as device performance is often compromised by intrinsic degradation processes that are masked in ambient environments. Here, we systematically investigate the stability of inverted PSCs with the architecture ITO/PTAA/$MAPbI_3$/PCBM/AZO/Ag under controlled atmospheres, continuous illumination, and thermal stress. Under nitrogen (N_2) in a glovebox, devices preserved >95% of their initial power conversion efficiency (PCE ≈ 16.5%) over 250 hours at 60 °C, with minimal decline in short-circuit current density (Jsc) and stable open-circuit voltage (Voc). In contrast, devices subjected to high vacuum (10^{-4} mbar) under identical conditions exhibited accelerated degradation, with PCE dropping to 12% within 250 hours and further plummeting to 3% after 95 hours under continuous 520 nm LED illumination (20 mW cm^{-2}). The sharp decline in Jsc and catastrophic fill factor (FF) losses indicate interfacial deterioration, electrode corrosion, and enhanced trap-assisted recombination. Intriguingly, while Voc remained largely unaffected, the divergence in FF evolution between nitrogen-stored and vacuum-stressed devices reveals distinct charge extraction limitations under different environments. These findings underscore the critical role of vacuum and thermal stress testing in isolating intrinsic failure pathways—including ion migration, perovskite lattice instability, and Ag electrode corrosion—that are otherwise suppressed by extrinsic factors such as oxygen and moisture. Our results highlight the urgent need for robust barrier strategies and interface engineering to suppress ion migration and stabilize charge transport layers, thereby enabling perovskite solar cells to meet the rigorous reliability demands required for real-world deployment. This work offers a promising strategy for designing vacuum-resilient OHP solar cells, with strong implications for space-based photovoltaic applications.

Introduction

Nowadays, perovskite solar cells have achieved power conversion efficiencies (PCE) surpassing >25% [1]. Among emerging photovoltaic materials, metal halide perovskites have garnered significant attention due to their outstanding optical and electronic properties [2]. These materials adopt the general formula ABX_3, where A represents a monovalent cation such as methylammonium (MA^+), formamidinium (FA^+), or cesium (Cs^+); B denotes a divalent metal cation commonly lead (Pb^{2+}) or tin (Sn^{2+}); and X corresponds to a halide anion—typically iodide (I^-), bromide (Br^-), or chloride (Cl^-) [3,4] This versatile composition enables fine-tuning of the perovskite's structural and electronic characteristics, facilitating optimization for photovoltaic applications. Obtaining high-quality perovskite films with optimal characteristics of generation and transport of photogenerated charge carriers is a serious problem [5]. In laboratory conditions, perovskite films are usually formed by spin-coating of a precursor solution and followed by heat treatment [6]. As a result, polycrystalline films are formed and from this point of view, control of morphology, grain size and substrate coverage level will be of decisive importance for the optoelectronic characteristics of the perovskite absorber [7]. Publications indicate that when exposed to water molecules and temperature, single-cation perovskite $CH_3NH_3PbI_3$ degrades to its original components, such as HI, CH_3NH_2, CH3NH3I and PbI_2 [8]. Additionally, perovskite decomposition has secondary effects, such as the deterioration of functional components in the solar cell caused by the corrosive nature of the decomposition products [9]. In particular, the corrosion of metal electrodes such as Ag or Al has been recognized as a significant concern [10]. Without effective strategies to address these reliability challenges, the widespread adoption and commercialization of organo lead halide perovskite technology could be severely hindered. Planar PSCs with a sandwich structure have two major architectures (p–i–n and n–i–p). Among all types of PSCs, the device based on (p-i-n) configuration is attracting more attention thanks to its negligible hysteresis at different scan rates or light intensities. Many p-type semiconducting materials have successfully been applied on PSCs as HTLs, such as spiro-OMeTAD [11-13], PTAA [14,15], PEDOT:PSS [16,17], and NiOx [18-20]. Among the diverse hole materials, poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine [PTAA, $(C_{21}H_{19}N)_n$] has become a popular candidate. Among diverse hole transport materials, poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine (PTAA) is one of the most promising candidates due to its ease of fabrication, transparency to visible light, mechanical flexibility, conductivity, and stability. The accumulation of excess PbI_2 at the interface between the perovskite layer and the hole transport layer reduces device performance when exposed to vacuum or nitrogen environments [21]. K.O. Brinkmann et al. demonstrated that integrating a bilayer electron extraction interlayer comprising AZO and ALD-deposited Oxidlayer beneath the Ag electrode in p-i-n structured PSCs substantially enhances device stability. This interfacial barrier effectively suppresses moisture ingress, prevents the escape of volatile perovskite decomposition products, and inhibits electrode corrosion, collectively enabling a T_{60} lifetime of 300 hours under continuous light and thermal stress at 60 °C [22]. In particular, single-cation perovskites containing volatile species such as MA^+ and MAI are prone to ion migration and material loss, especially under vacuum, thermal, and light-induced stress. We study stabilty performance of based on MAPbI3 solar cell with inverted under high-vacuum conditions was assessed, confirming that the devices maintained structural and functional integrity without significant degradation. This indicates strong resistance to vacuum-induced stress. The cells were tested under realistic operational conditions, including continuous light exposure and elevated temperatures. These stress factors provided insights into long-term behavior and potential degradation pathways. The results indicate that incorporating an oxide layer is essential for device stability. This layer effectively mitigates ion migration under continuous light exposure (520 nm LED, ~20 mW/cm²) and prevents electrode degradation at elevated temperatures in high vacuum conditions (10^{-4} mbar). The demonstrated device structure was ITO/PTAA/$MAPbI_3$/PCBM/AZO/Ag.

Metal halide perovskite solar cells (PSCs) have emerged as a promising photovoltaic technology due to their high power conversion efficiencies (PCEs), low-cost fabrication, and compatibility with scalable processing methods. Inverted device architectures, such as ITO/PTAA/$MAPbI_3$/PCBM/AZO/Ag, have attracted particular attention owing to their reduced hysteresis, low-temperature processing, and potential for tandem integration. Despite rapid progress, however, the long-term operational stability of PSCs remains a critical bottleneck to commercialization. While much of the observed instability in PSCs arises from extrinsic factors such as oxygen, moisture, and UV exposure, intrinsic degradation pathways—including ion migration, lattice instability, and interfacial deterioration—remain insufficiently understood. Controlled stability studies under inert or vacuum environments are therefore essential to decouple these intrinsic mechanisms from extrinsic degradation. In particular, understanding the role of charge transport layers, electrode reactivity, and ion dynamics under thermal and illumination stress is key to guiding materials and device design strategies. Previous reports have demonstrated that the choice of environment strongly influences degradation pathways: devices exposed to ambient conditions often fail rapidly due to moisture ingress and oxygen-induced reactions, whereas those stored in inert atmospheres degrade more slowly. However, fewer studies have systematically compared device behavior under nitrogen and vacuum, where differences in gas composition, pressure, and electrode stability may reveal distinct intrinsic degradation processes. In this work, we investigate the stability of inverted $MAPbI_3$-based PSCs with the architecture ITO/PTAA/$MAPbI_3$/PCBM/AZO/Ag under two controlled conditions—nitrogen and high vacuum—at elevated temperature and under continuous illumination. By isolating intrinsic degradation pathways, we provide insights into the roles of ion migration, interfacial contact deterioration, and silver electrode corrosion in governing long-term stability. Our findings highlight environment-dependent degradation mechanisms and underscore the importance of barrier strategies and interface engineering to suppress intrinsic instabilities, offering pathways toward extending the operational lifetimes of PSCs for practical applications.

Solar cell fabrication

Our device structure which is ITO/PTAA/MAPbI3 (+5%PbI2)/PCBM/AZO/Ag. The active area of 3.14 mm^2 was defined by patterned photoresist layer on ITO-coated glass. Indium tin oxide (ITO)-coated glass substrates were cleaned sequentially in deionized water ,acetone, isopropanol at drayed with N2 gas each part followed by UV-ozone treatment for 15 minutes. The hole transport layer was formed by spin-coating poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine] (PTAA) solution (5 mg/mL in toluene) at 4000 rpm for 30s, followed by annealing at 120°C for 10 minutes. The perovskite precursor solution was prepared by dissolving methylammonium iodide (MAI) and PbI_2 in a 1:1.05 molar ratio (with 5% excess PbI_2) in N,N-dimethylformamide (DMF) and N-Methyl-2-pyrrolidone (NMP) (7:3 rate).100 mg/mL thiourea was added into the precursor solution. Active layer were deposited by spin-coating at 2000 rpm for 120 s. During after the start 15 seconds, a nitrogen gas stream was applied to induce rapid crystallization (gas quenching). Films were then annealed at 100°C for 10 minutes. Subsequently, [6,6]-phenyl C_{61} butyric acid methyl ester (PCBM, 25 mg/mL in chlorobenzene) was spin-coated as the electron transport layer and dryed at room temperature. AZO is diluted 1:2 volume ratio with isopropanol and 100µl were spin coated at 4000 rpm for 20s followed by an annealing step at 80°C for 70 min. As an electrode, 100 nm of silver were thermally evaporated in high vacuum.

Results and Discussion

Moisture and oxygen induced degradation

When perovskite solar cells operate in a nitrogen atmosphere, only lattice shrinkage is observed, which contributes to enhanced stability of the devices. In contrast, under vacuum conditions, these issues are more pronounced. Furthermore, the migration of excess PbI2 to the interface between the perovskite layer and the hole transport layer occurs in both nitrogen and vacuum environments. This migration significantly compromises the device's efficiency by disrupting the interface, ultimately leading to reduced overall performance. The findings suggest that nitrogen plays a crucial role in stabilizing the perovskite solar cells by mitigating harmful degradation processes [21]. As indicated earlier, moisture is a major factor that leads to the degradation and destabilization of perovskite solar cells (PSCs). The degradation process of perovskites involves a chemical reaction where moisture acts as a catalyst. Perovskite materials decompose when exposed to moisture, mainly because they are highly susceptible to water and prone to hydrolysis due to their polar characteristics.

Stability of a perovskite solar cell with the architecture ITO/PTAA/MAPbI3/PCBM/AZO/Ag was systematically evaluated under nitrogen (N_2) conditions within a glovebox to eliminate extrinsic degradation factors such as oxygen and moisture. This controlled environment allowed isolation of intrinsic material and interfacial stability mechanisms. The device was subjected to continuous monitoring over 55 days, with periodic measurements of its photovoltaic parameters, including power conversion efficiency (PCE) and fill factor (FF), under standard AM 1.5G illumination (Fig 1.). The device exhibited remarkable stability during the first 30 days, with no measurable degradation in PCE or FF. The long-term stability of perovskite solar cells (PSCs) is closely linked to their power conversion efficiency (PCE), as devices with higher PCEs often exhibit more optimized interfaces, superior crystallinity, and better-controlled fabrication processes—factors that inherently contribute to improved operational durability. A gradual decline in PCE (1.5–2% reduction) was observed, likely attributed to minor ion migration or defect accumulation within the perovskite lattice. Concurrently, the fill factor (FF) decreased by~10%, indicating emerging resistive losses or charge recombination at the carrier transport layers (e.g., PTAA/perovskite or PCBM/AZO interfaces) .A rapid deterioration of performance occurred in the final 5 days: PCE plummeted from 15% to 10% (a 33% relative loss), signaling catastrophic failure of the active layer or interfacial contacts. FF collapsed from 65% to 55%, implying severe carrier extraction inefficiency or shunting pathways.

The obtained data confirm the pattern of similar studies conducted in the surrounding atmosphere with different humidity, with the exception of slow degradation in our conditions due to the low concentration of water and oxygen of no more than 0.1 ppm.

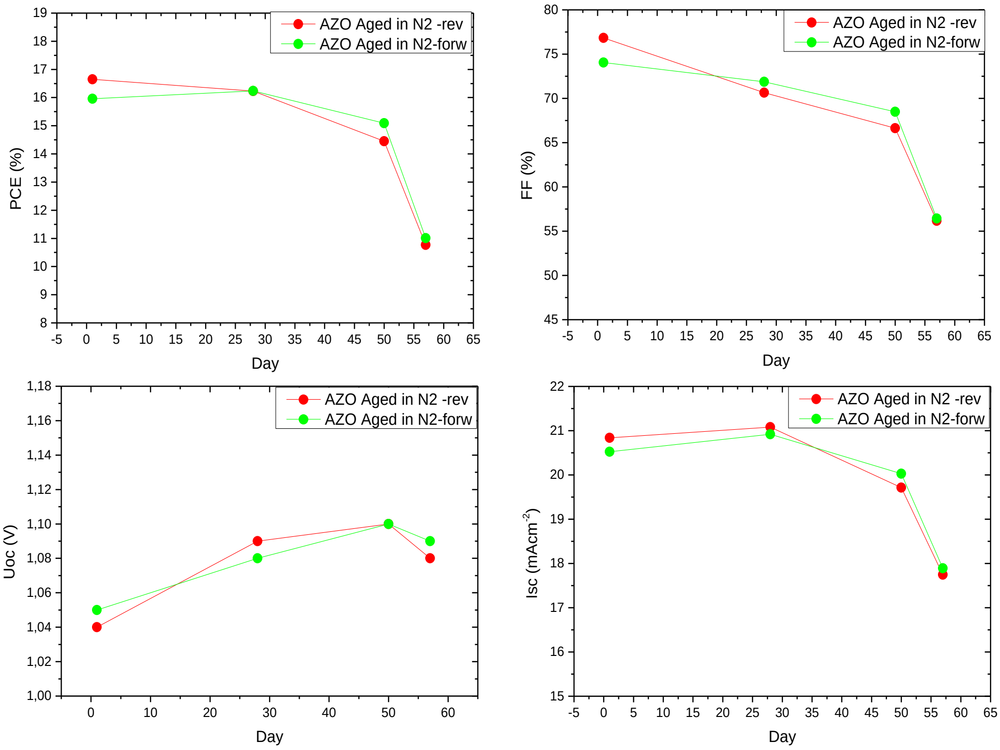

Figure 1. *Stability of aged solar cell in nitrogen atmosphere.*

Stability at elevated temperatures

Temperature is an inevitable stressor in photovoltaic applications, as sunlight radiation includes infrared energy that heats the materials. According to international standards [23], heating between 60 and 85°C is considered normal during operation. However, it is important to have a margin for higher temperature endurance to ensure the material can with stand conditions in hot climates or when integrated into technologies that may generate additional heat during operation [24,25]. Structural changes in the perovskite layers were observed, which could be detected by measuring the material's conductivity using conductive atomic force microscopy. As the temperature increased, the conductivity of the perovskite rapidly decreased, likely due to the formation of PbI2, a wide bandgap material. The temperature-induced decomposition process is represented by the following equation (1)

$$CH_3NH_3PbI_3 \rightarrow CH_3NH_2\,(g)\uparrow + HI(g)\uparrow + PbI_2(s) \qquad (1)$$

The stability of the ITO/PTAA/$MAPbI_3$/PCBM/AZO/Ag perovskite solar cell was evaluated under two different conditions: in a nitrogen atmosphere (inside a glove box) and under vacuum, both placed on a hot plate maintained at 60 °C (Fig 2). The results indicated that the power conversion efficiency (PCE) degraded more significantly under vacuum compared to the nitrogen environment. Specifically, under vacuum conditions, the PCE dropped from 16% to approximately 12% after 250 hours. In contrast, under the nitrogen atmosphere, the PCE showed a smaller decline, decreasing from 16.5% to just below 16% over the same time period. The open-circuit voltage (Voc) remained nearly constant in both environments, suggesting that the built-in potential was not significantly affected. However, the short-circuit current density (Jsc) decreased in both cases—from 21 mA/cm² (in nitrogen) and 20 mA/cm² (in vacuum) down to a similar final value of 17.5 mA/cm². Interestingly, while the fill factor (FF) increased under the nitrogen atmosphere, it decreased when the device was stored in vacuum, pointing toward different degradation mechanisms affecting charge extraction and transport under each condition.

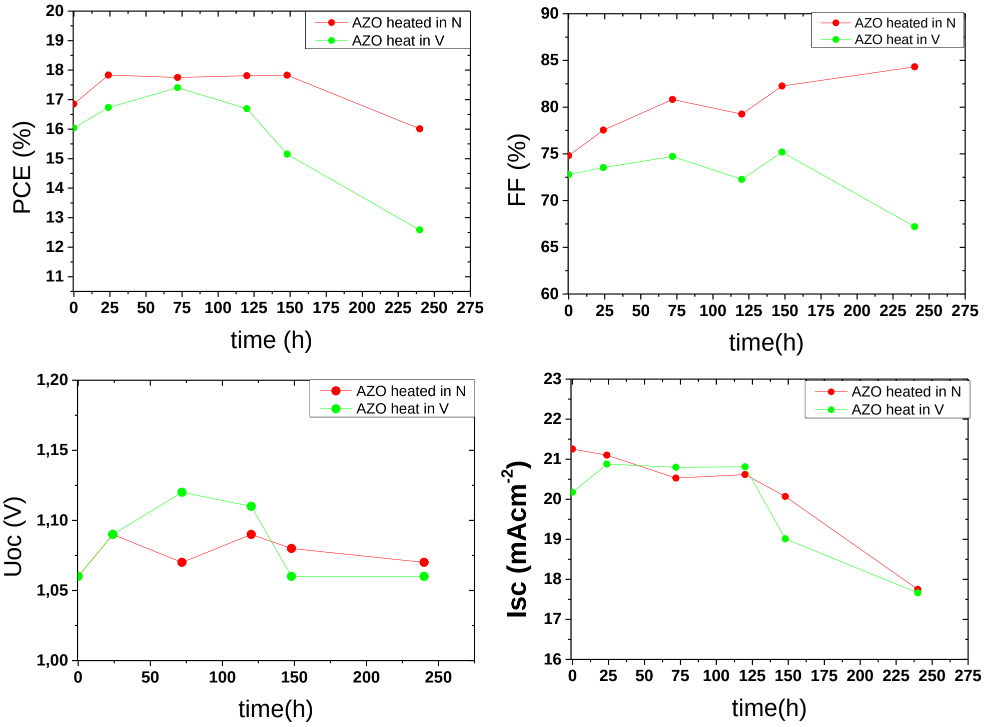

Figure 2. *Stability upon storage at 60 °C in nitrogen atmosphere and vacuum condition*

Illumination induced degradation

Effective sealing can reduce the impact of environmental factors like oxygen and humidity. However, for a semiconductor to last as a solar material for more than twenty-five years, it needs to be naturally resistant to light exposure and other factors that cause damage. Illumination is an essential factor in generating a photovoltaic response in solar cells. Verifying that a material maintains stability after prolonged exposure to light is generally the initial step in evaluating the suitability of the chosen active material for use in photovoltaic devices [26,27,28].

Table 1

Stability of PCS under light in nitrogen atmosphere.(AZO)

Time (hours)	PCE(%)	FF(%)	Uoc (V)	Isc(mA/cm2)
0	15,80207	69,66288	1,08	21,00336
72	16,36608	73,74574	1,11	19,99331
120	16,9233	75,80481	1,1	20,29531
144	17,75405	76,16817	1,13	20,62745
168	16,75866	73,9759	1,13	20,04798

From Table 1. given datas that the ITO/PTAA/MAPbI$_3$/PCBM/AZO/Ag perovskite solar cell architecture demonstrates exceptional operational stability under accelerated light-stress conditions, the device maintained and even improved its photovoltaic performance over a 168-hour period. Based on the provided data and supported by recent literature, the ITO/PTAA/MAPbI$_3$/PCBM/AZO/Ag perovskite solar cell exhibits remarkable operational stability under continuous illumination. Subjected to monochromatic 520 nm LED light at 20 mW/cm^2 in a nitrogen atmosphere for 168 hours, the device maintained and even improved its photovoltaic performance. The PCE increased from 15.80% at initial to a peak of 17.75% at 144 hours, before slightly decreasing to 16.76% at 168 hours. This enhancement is primarily attributed to improvements in the fill factor (FF) and open-circuit voltage (Voc). Such behavior is characteristic of the light-soaking effect, where illumination leads to defect passivation and enhanced charge transport . The FF improved from 69.66% to 76.17% over the 144-hour period. This suggests enhanced charge extraction and reduced recombination losses, likely due to the passivation of trap states at the perovskite/PCBM interface. Voc increased from 1.08 V to 1.13 V, indicating a reduction in non-radiative recombination pathways. This enhancement is consistent with improved interfacial quality and energy level alignment between the perovskite and charge transport layers. Jsc remained relatively stable, fluctuating slightly from 21.00 mA/cm^2 to 20.04 mA/cm^2. This stability suggests consistent photo-generation and minimal optical or morphological degradation within the perovskite layer.

Extended Analysis of J–V Characteristics of MAPbI$_3$ Devices under Operational Stress in vacuum: Now it is interesting to follow the changes in the parameters under study when introducing an additional electron-conducting layer in the form of SnOx into the cell structure.The device architecture employed was ITO/PTAA/MAPbI$_3$/PCBM/AZO/Ag.To evaluate the operational stability of MAPbI$_3$-based perovskite solar cells, J–V characteristics were systematically investigated under continuous illumination (LED, 520 nm, intensity = 20 mW/cm^2) for 4 days in a high-vacuum environment (10^{-4} mbar). The device exhibited significant degradation in performance over time(Fig-3). Initially, the PCE declined from 17% to 15% during the first 50 hours, followed by a steep decrease to 3% in the subsequent 45 hours. This dramatic loss of efficiency is indicative of intrinsic material or interfacial instability under vacuum and illumination stress, likely due to increased trap-assisted recombination, morphological degradation, or interface deterioration. The device saw a reduction in Jsc from 20 mA/cm^2 to 18 mA/cm^2 within the first 50 hours, followed by a substantial drop to 2 mA/cm^2 in the remaining time(Fig-3). This trend indicates a severe loss in photo-generated charge collection, possibly due to deterioration in perovskite crystallinity or charge transport layers. Interestingly, the FF of the device increased from 73% to 75%, and further to 82%, which may seem counterintuitive. This apparent improvement might be attributed to reduced current (Jsc) resulting in less series resistance effects or artifactually improved FF due to lower photocurrent, a phenomenon sometimes observed in degraded cells.

No Recovery in Devices:, devices did not exhibit any observable recovery after the 12-day period. The performance metrics remained at degraded levels post-stress, indicating permanent or non-reversible damage under operational stress.

The lack of recovery in the solar cell configuration may be attributed to irreversible interfacial degradation, metal diffusion (e.g., Ag migration), or the formation of deep trap states that are not passivated in the absence of Oxidlayer. This comparison highlights the importance of interlayer engineering in achieving not just long-term operational stability, but also self-healing capacity under realistic operating conditions.

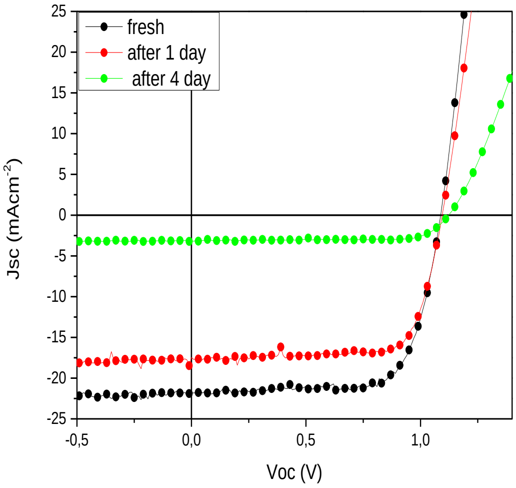

Figure 3. J-V characteristics of a $MAPbI_3$ device under operation with light (LED, 520nm/P=20mW/cm^2) in harder vacuum conditions of 10^{-4} mbar for 4 days. ITO/PTAA/MAPbI3/PCBM/AZO/Ag

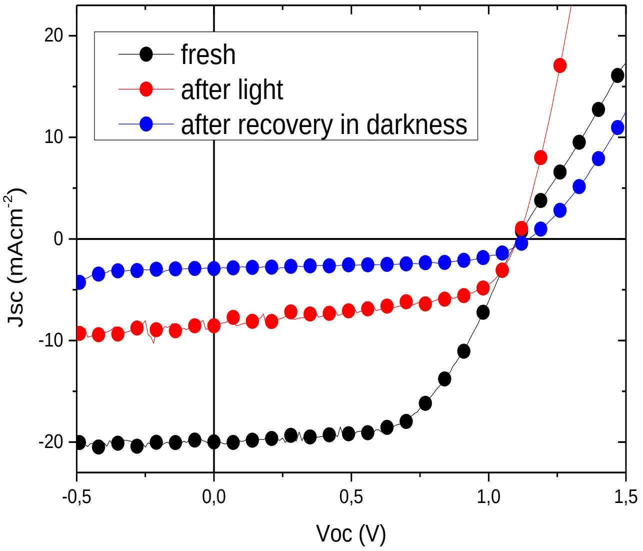

Figure 4. J/V characteristics of perovskite solar cell, non-stressed (fresh), stressed for 96 hours (light), and subsequently recovered in in darkness at room temperature for 12 days (in the glove box).

Photographic Evidence of Electrode Corrosion under Thermal Stress in High Vacuum

We observed that thermal stress in vacuum can cause the top electrode to shift from a metallic silver hue to a brownish-black coloration (Fig-5), rendering the devices non-functional - an effect typically indicative of silver iodide formation, degradation reaction induced by ion migration.

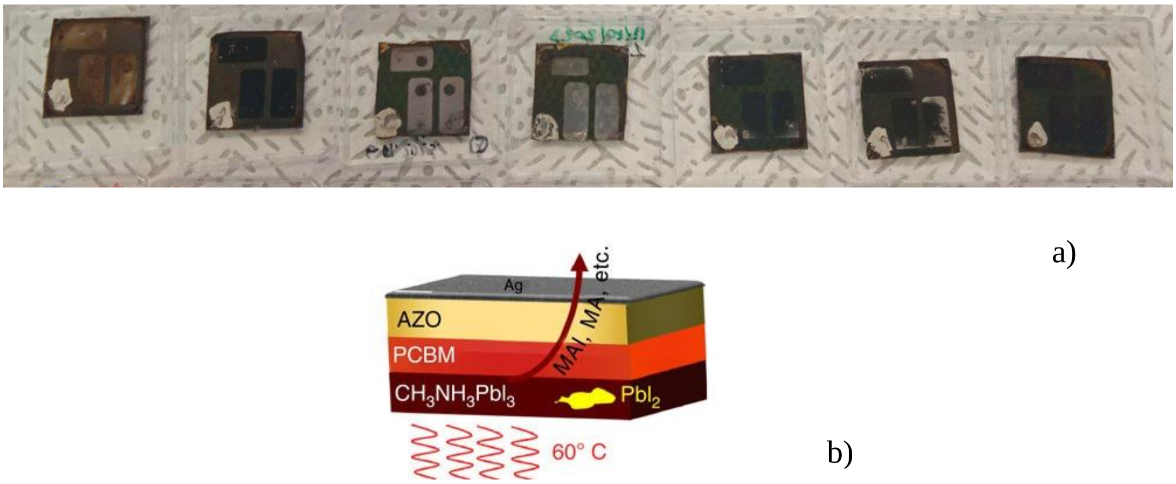

***Figure 5.** a) Photographs showing the colour change (the corrosion of metal electrode) upon thermal stress under the same high vacuum conditions (10^{-4}mbar; 60 ^{0}C). MAPbI$_3$ device .* ***b)** Schematic illustration of suppressed degradation pathways, adapted from Ref. 22.*

We conducted thermal stability tests under high vacuum conditions (10^{-4} mbar at 60°C). The study focused on the device architecture with ITO/PTAA/MAPbI$_3$/PCBM/AZO/Ag. Photographs taken after the thermal stress exposure clearly reveal a significant difference in electrode stability. In the device, visible darkening of the silver (Ag) electrode was observed, indicating corrosion or degradation, likely due to ion migration and interaction between Ag and halide species diffusing from the perovskite layer under heat and vacuum (Fig 5a). Methylammonium (MA) and methylammonium iodide (MAI) cations are inherently volatile, which increases the likelihood of material degradation and loss when subjected to harsh environmental conditions. Since phase separation is unlikely in a single-cation perovskite system, ion migration–driven material loss is identified as the most probable mechanism underlying this degradation. We tried to study that the degradation mechanism under vacuum conditions mirrors the instability previously observed at elevated temperatures, where material loss played a critical role .

Conclusion

The stability of ITO/PTAA/MAPbI$_3$/PCBM/AZO/Ag perovskite solar cells was systematically investigated under controlled conditions to disentangle intrinsic degradation from extrinsic effects. Under inert nitrogen, devices maintained excellent operational stability for the first 30 days, showing negligible loss in PCE(16%) and FF(73%). Gradual degradation emerged thereafter, with a 1.5-2% PCE drop and ~10% FF reduction, followed by catastrophic failure in the final days, marked by a sharp efficiency decline (15% → 10%) and severe carrier extraction losses. Comparative testing at 60°C further revealed environment-dependent degradation pathways: while nitrogen-stored devices retained nearly all of their PCE over 250 hours, vacuum-stressed cells degraded more severely (16% → 12%), despite stable Voc in both cases. Declines in Jsc and divergent FF behavior point to distinct charge transport and interfacial failure mechanisms under the two environments. Overall, these findings confirm that eliminating oxygen and moisture suppresses extrinsic degradation, yet intrinsic processes-such as ion migration, interfacial deterioration, and electrode instability-still govern long-term device failure. To extend operational lifetimes toward practical deployment, advanced barrier layers, interface engineering, and ion-migration mitigation strategies will be essential.

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