



Recent Advances in Addressing J-V Curve Hysteresis in Perovskite Solar Cells

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ABSTRACT

Despite their incredible potential as replacements for traditional silicon cells, perovskite solar cells (PSC) are prone to J-V curve hysteresis which prevents accurate performance measurement. This paper synthesizes recent research relevant to understanding this issue. The primary causes are identified as ion migration, charge trapping, and ferroelectric effects within the perovskite. Studies have shown that doping solutions to improve crystal quality and defects, in addition to interface engineering to modify or replace the charge transport layers, show effective results. Furthermore, new techniques can help estimate hysteresis more accurately. While these solutions are remarkable, further real-life conditions testing, as well as economic and environmental simulations remain to be done to assess the scalability of production of these cells with new materials. Advances in hysteresis research are critical to the successful exploitation of PSCs in the future.

Keywords: Perovskite solar cells, Hysteresis, Ion migration, Charge trapping, Doping, Interface engineering, PSC: Perovskite Solar Cells, PSK: Perovskite, PCE: Power Conversion Efficiency, FF: Fill Factor, V_{oc} : Open-circuit Voltage, J_{sc} : Short-circuit Current, P_{in} : The input from the sun, CPE: Constant Phase Element, HTL: Hole Transport Layer, ETL: Electron Transport Layer, CTL: Charge Transport Layer.

INTRODUCTION

Thanks to their easy band gap tunability, low charge recombination rates [1], scalability, flexibility and mild environmental threat compared to traditional silicon alternatives [2], coupled with a superior power efficiency of 34.6% [3], PSC are a prime candidate to replace traditional solar cells. They face, nevertheless, several limitations, among which are stability issues under elevated humidity and temperature conditions, concerns about lead toxicity [1] and hysteresis leading to commercialization issues.

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This hysteresis impacts performance evaluation and stems mainly from ion migration and interfacial ion accumulation [4]; it can be classified as either capacitive or inductive and appears on current-voltage curves [5]. Comprehending this phenomenon is crucial for precise performance evaluation of PSC, as it affects power efficiency measurement.

This review aims to investigate the main causes of hysteresis in PSCs and provide a concise synthesis of how recent research has advanced our understanding of this problem, indicating future research possibilities.

FUNDAMENTAL MECHANISMS BEHIND J-V HYSTERESIS

A variation in the shape of the resulting J-V curve when performing a voltage scan in the forward direction and the curve obtained when performing it in the reverse direction is known as hysteresis. Thus, the severity of it is influenced by the direction of the sweep and the sweep rate. The most common form of hysteresis is where reverse scanning yields higher electrical results and efficiency, whereas in inverted hysteresis, the forward scan shows better results. Interestingly, inverted hysteresis can serve as a diagnostic tool for understanding cell properties [6]. The efficiency of a PV cell is defined as $PCE = \frac{FF * V_{oc} * J_{sc}}{P_{in}}$ where FF represents the fill factor, V_{oc} the open-circuit voltage, J_{sc} the short-circuit current and P_{in} the input from the sun. By affecting parameters like FF, V_{oc} , J_{sc} and operational stability, hysteresis influences the efficiency [7]. In this section, we talk about the causes leading to hysteresis.

One of the primary causes of hysteresis is ion migration [8] [4]. This phenomenon involves the movement and accumulation of ionic species, particularly organic cations, at the interfaces [4] [9]. Variety of studies support this theory, such as an experiment by Kumar et al. [10] showing that by cooling the perovskite down from 300 K to below 180 K in the dark where there is no luminosity influencing the results, hysteresis decreased as ion migration is a thermal process. While often considered detrimental [11], ion accumulation can also enhance device performance by increasing built-in potential and open-circuit voltage [12]. Hysteresis manifests in two main forms: capacitive and inductive, depending on the dominant equivalent circuit component [5] [13]. Overall, understanding and controlling ion migration is crucial for producing highly efficient and stable PSCs [13].

Furthermore, charge trapping and de-trapping can also cause hysteresis, where charge carriers are captured and released by defects [14], [15]. Device performance is negatively impacted by this process as it prevents trapped charges from contributing to the electrical current. The severity of the hysteresis scales with the number of traps as a higher number leads to a higher hysteresis [16][17].

From an electrical perspective, the combined effect of ion migration and charge trapping is an anomalous capacitive effect. While not a physical cause, it is an electrical symptom resulting from the physical ones, as shown by Lee et al. [18], which indicates that a reduction in “capacitance” results in reduced hysteresis. This issue was considered from a theoretical perspective by Hernández-Balaguera et al. [19] [20] by using a new “theoretical framework” that describes the electrical behavior of PSC. It reveals increased hysteresis when “non-ideal capacitive” effects are considered.

Another perspective explains hysteresis as a consequence of ferroelectric polarization, which is the ability of a material to show spontaneous polarization whose orientation can be altered with an impactful external electric field, which causes the formation of alternating polar domains[21][22]. This behavior is a cause of hysteresis [23] as this reorientation affects the fill factor, power conversion efficiency [24].

RECENT ADVANCEMENTS IN UNDERSTANDING J-V HYSTERESIS

This chapter details the recent advancements in understanding and reducing hysteresis.

Assessment of Hysteresis

Perovskite solar cells behave differently depending on how quickly or slowly they are measured because of hysteresis. Following their “theoretical framework” in the 2020 paper linking non-ideal capacitive effects to hysteresis, Hernández-Balaguera et al. [25] proposed an experimental protocol involving an impedance spectroscopy measurement at different voltages, this data is then introduced into an “electrical circuit model that includes a Constant Phase Element (CPE)” to extract different parameters. They are then used to calculate, using different equations, the optimal scan rate. The results are hysteresis-free measurements verified by scans at different speeds. In a most recent paper, Hernández-Balaguera et al. [26] developed an improved four-step protocol that allows for hysteresis-free measurement. It involves performing a fast initial scan followed by transient response measurements over a fraction of a second. This information is then introduced in specific formulas to deduce an optimal scan rate leading to a hysteresis-free J-V curve and reducing measuring time by at least half. To reduce the toxicity of PSCs, tin-lead can be a good alternative. To accurately measure these cells and eliminate hysteresis, Zhang et al. [27] employed a series of tests under varying temperatures, humidity, measurement steps, measurement delay times and bias voltages to estimate power conversion and hysteresis. They concluded that with a measurement time no longer than 30 s, a temperature below 25 °C and under 10% relative humidity if the latency is superior to 0.1 s with a measurement step inferior to 10 mV/s, a reverse voltage bias of -0.2 V and a forward one of 120% V_{oc} , the most accurate performance parameters are obtained.

A different study by Veurman et al. [16] establishes a method of estimating the roles of ion migration and charge trapping in causing hysteresis by measuring under specific conditions of low light (0.01 sun) coupled with a fast measuring speed (>100 V/s) and observing the appearance of a diffusion capacitive effect which indicates a dominance of charge traps. In contrast, a lack of this effect indicates a dominance of ion migration. This method was achieved by comparing simulations under the assumption that slow-shallow traps (SST) and ion vacancies are culprits. A distortion in the graph appeared when measuring at speeds greater than 100 V/s and low light, with the distortion being more pronounced as the speed increased when SSTs are the culprit. This distortion, called diffusion capacitive effect, does not appear in the second assumption.

Doping Solutions

Due to the strong link between charge transport imbalance and the hysteresis phenomenon, doping has been investigated as a solution to enhance device capabilities. Doping either the interfaces or the perovskite layer (PSK layer) itself allows for a reduction in ion migration and defects, in addition to improving charge transport. Aluminum acetylacetonate doping on the

PSK layer was tested as a solution by Wang et al. [28]; it was shown to reduce micro stain, thus improving crystal quality, which resulted in a reduction of non-radiative recombination, leading to a 19.1% PCE with minimal hysteresis. Potassium iodide was shown by Son et al. [29] to significantly reduce hysteresis in both pure and mixed perovskite, with the reduction being more important in the latter, through the elimination of Frenkel defects and a reduction in trap states. This result is achieved due to potassium ions occupying the interstitial sites, thereby stabilizing the crystal. Weiji Li et al. [30] achieved similar results by adding potassium iodide to a MAPbI_3 perovskite and dissolving it. They then spin-coated perovskite thin films with it, ensuring an even distribution of KI within the perovskite. This resulted in higher-quality crystals with fewer defects and provided more than defect passivation, as it changed the perovskite from N to P-Type conductivity, lowering the valence band's upper limit and the conduction band's lower limit, therefore improving power conversion efficiency from 15.15% to 20.66% and eliminating most of the hysteresis. Moreover, numerical simulations show an improvement in open-circuit voltage attributed to a larger quasi-Fermi energy level difference. The precursor factor of $\text{CH}_3\text{NH}_3\text{PbI}_3$, defined as the ratio of PbI_2 to MAI, was altered by Kari et al. [31] from 0.30 to 0.80, where hysteresis factor decreased from 19.5% to 8.87%, while it increased when going above 0.80 reaching 29% at 1.70. These precursor factor changes also altered the between P-Type at 0.30, intrinsic, and N-type at 1.70. These results are, however, only simulations and lack experimental confirmation. Focusing on PbI_2 too, Chen et al. [32] used in-situ X-ray diffraction (XRD) to understand the reaction between PbI_2 and organic salts. They found that the reaction formed $\text{FAPbI}_{3-x}\text{Br}_x$, and extended heating at 4 minutes or more resulted in the formation of PbI_2 crystals from the leftover PbI_2 . These crystals helped reduce hysteresis from 23.55% to 0.50% and improved efficiency thanks to a reduction in defect density and ion migration. Mahmoudi et al. [33] opted to create a new PSC with a "MAPbI₃-xCl_x/NiO-graphene photoactive composite and NiO interface layer" between the ETL and perovskite absorber. This new PSC reached 95.2% of the theoretical maximum photocurrent density and a 20.8% efficiency with no hysteresis. The perovskite absorbing layer was doped using ammonium cations (EA^+) by Khan et al. [34]. EA^+ diffused seamlessly into lead iodide (PbI_2) and improved PSC performance due to less dense defect states. The PSC reached 21.32% PCE in the reverse scan and 21.09% in the forward scan, resulting in minimal hysteresis. Zhang et al. [35] proposes that polymers can also be used as additives to the PSK layer to improve capabilities and hysteresis.

Interface Engineering

The power conversion of PSCs is also determined by the properties of the interfaces where the layers connect. The most used electron transport layer (ETL) and hole transport layer (HTL) layers in PSCs, Tin Oxide (SnO_2) and Spiro-OMeTAD, respectively, can have a role in reducing cell efficiency and thus causing hysteresis. To address this issue, the detrimental hydroxyls from the SnO_2 layer and lithium ions from the Spiro-OMeTAD layer were simultaneously passivated and immobilized, respectively, by Li Yin et al. [36] using 3,5-bis(fluorosulfonyl)benzoic acid (FBA) as an interfacial modifier at the SnO_2 surface. This resulted in reduced interfacial traps and improved energy alignment, raising the PCE to 24.26% without hysteresis, in addition to enhancing moisture resistance and operational stability. The same approach was used by Bi et al. [37] with oteracil potassium (OP) as a layer between SnO_2 and methylammonium-free perovskite ($\text{Rb}_{0.02}(\text{FA}_{0.95}\text{Cs}_{0.05})_{0.98}\text{PbI}_{2.91}\text{Br}_{0.03}\text{Cl}_{0.06}$) layer, reducing oxygen vacancy in SnO_2 and passivating Pb^{2+} . While this allows the PCE to reach 22% with no hysteresis and notable

humidity stability, it does not reach a conclusion as to whether the reduction in hysteresis stems mainly from the contact improvement between the layers or the PSK layer passivation. On the other hand, phenyl-C61-butyric acid methyl ester (PCBM) was layered on top of the PSK layer by Zhong et al. [38] and was shown, using wide-field photoluminescence imaging microscopy, to neutralize iodine-related defects thus reducing hysteresis. This is due to PCBM molecules being mobile and migrating to the PSK layer, passivating defects. Furthermore, while chemical modifications are important, the physical architecture of the ETL has been shown to impact hysteretic behavior directly. A study by Rana Yekani et al. [39] comparing planar TiO₂ with 3D scaffold-based architecture found that the flat interface led to a reduction in hysteresis by improving the fill factor (FF) over time with higher thickness sustaining the FF more in forward scans. A combination of capacitance-voltage profiling, drift-diffusion simulation, and experimental measurements were used by Regalado-Pérez et al. [40] to compare the traditional Spiro-OMeTAD HTL with CuSCN showed that the latter had caused less hysteresis due to more homogenous distribution of charges and the absence of Li⁺ and Co³⁺ ions leakage from Spiro-OMeTAD. However, Spiro-OMeTAD still had better PCE, showing that less hysteresis does not always lead to more efficient cells. After changing the precursor ratio, Kari et al. [31] altered the HTL and ETL layers proposing a structure of Cu₂O as HTL and ZnO as ETL; this further decreased the hysteresis factor to 4.24%, a 52.73% decrease from the reference structure. Ionic liquids can be a solution for more consistent PSCs as an addition to the absorbing layer or the interfaces. González-Juárez et al. [41] spin-coated tetrabutylphosphonium iodide (B₄PI) into the TiO₂ ETL at different concentrations at 5000 rotation per minute for 30 seconds, then warmed it up to 80°C for 5 minutes and allowed it to reach ambient temperature before the PSK layer was deposited. They found that at 0.3% concentration, performance was slightly reduced, but hysteresis improved from 17% to 11%.

Yinghui Wu et al. [17], on the other hand, spin-coated HBPC ((4'-pentyl-[1,1'-biphenyl]-4-yl) methanaminium iodide) into the PSK layer. It resulted in a PCE improvement from 20.04% to 22.07% and a hysteresis index of 9.0% at a concentration of 4 mg/mL of HBPC. Nickel oxide (NiO_x) is often used in PSCs as HTL. A study by Cheng et al. [42] aimed to improve the performance of these PSCs using three different layers, derived from "dibenzofulvene-bridged triphenylamine (TPA)", inserted between NiO_x and the PSK layer, TPAD, TPAO, and TPAS. The PSC using TPAS as a layer yielded the highest efficiency at 20.30% with very low hysteresis. Ambient-condition fabricated PSCs were given special attention by Amrollahi Bioki et al. [43], as they showed an improvement of 22.4% in efficiency, reaching 14.64 % PCE in the best-case and reduced hysteresis from 20.7% to 7.8%. This was done by inserting a thin layer of "HKUST-1 metal organic framework (MOF)" between the TiO₂ ETL and the PSK layer. Nair et al. [44] passivated low-cost carbon-based PSCs using acetic acid at 8% in a chlorobenzene anti-solvent during spin-coating and CaCl₂ at 0.6% molar ratio directly into the perovskite. They were tested using an accelerated aging test from 35 °C to 95 °C at 85% RH, which revealed that the PSCs treated with acetic acid displayed a diminution in hysteresis. Lee et al. [18] investigated the fabrication of TiO₂ under ambient air combined with thermal-assisted UV annealing at low temperatures, by spin-coating a titanium precursor solution onto an FTO substrate. They tested different temperatures, ranging from ambient temperature to 150 °C, and deduced that the best results were obtained at 125 °C, showing a PCE of 20.36% with reduced hysteresis. Temperatures ranging from 100 °C to 150 °C showed an improved PCE fill factor and less hysteresis. Mahmood et al. [45] attempted to replace traditional ETLs with hydrothermally

synthesized MoS₂ by electrospraying it onto FTO glass. These PSCs achieved 16.09% efficiency, compared to spin-coated MoS₂ at 13.79%, with negligible hysteresis. A defect post-passivating method, where the passivating material is applied after the PSK layer has been formed, was used to enhance the performance of PSCs as shown by Feng et al. [46]. MAPbI₃ films were passivated using triphenylphosphine oxide (TPPO) with this method, protecting them from moisture and oxygen degradation. These PSCs achieved 14.9% PCE with minimal hysteresis at 0.1 mmol/L concentration of TPPO. Aranda et al. [47] explored alkali metals as a potential solution to mitigate ion-driven interfacial recombination, which results in hysteresis. Li, Na, and K were each spin-coated onto the TiO₂ layer of a MAPbBr₃ PSC, respectively, and studied using impedance spectroscopy for comparison. Sodium (Na⁺) was found to be the most effective as it prevented ion migration due to methylamine compounds interacting electrostatically with sodium. Zhang et al. [48] used sodium citrate modified poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (SC-PEDOT: PSS) as “conductive adhesive”. It was mixed with a perovskite precursor ink and then spin-coated into the TiO₂ layer, improving crystallization, enhancing charge transport, and passivating defects. The device reached 19.1% efficiency with very low hysteresis and had superior humidity resistance.

In a review of carbon-based charge transport layers (CTL), Ali et al. [49] emphasized that carbon-based CTLs reduce hysteresis in PSCs in addition to improving PCE and stability. In a similar trend, bidentate ligands (a type of molecule with two donor atoms) were shown in a review by Ugwu et al. [50] to improve efficiency and reduce hysteresis by passivating defects either in the perovskite cells or the interfaces. Zhang et al. [35] proposed that polymers can be used to improve PSC performance, including hysteresis, by passivating or acting as CTLs.

CONCLUSION

Thanks to the high potential of PSCs as an alternative to current mainly used technologies, J-V hysteresis is a highly investigated issue. This irregularity is the result of several mechanisms, such as ion migration, charge trapping, and ferroelectric polarization.

Analysis of recent research has shown that innovative protocols have been developed to help achieve measurements with no hysteresis by optimizing scan rates and specific conditions. It also highlighted physical solutions to this phenomenon by altering the PSK layer itself or its interfaces and the layers, linking them, through doping and interface engineering. Some of the proposed solutions have limited reproducibility or are done under idealistic conditions, which puts into question long-term performance. They also in certain cases focus on hysteresis reduction, sacrificing efficiency. The newly introduced materials, while they improve hysteresis, may introduce new challenges such as toxicity, stability issues, or manufacturing issues which could increase the price of these cells. While understanding of hysteresis is improving, continued research in addressing defects in the PSK layer and the interfaces without sacrificing efficiency, as well as, increased experimental and long-term tests remains critical to a stable, efficient, and, commercialized perovskite solar cell.

Declarations

Availability of Data and Materials

All data analyzed during this study are included in the published articles cited. No new datasets were generated or analyzed in this review.

Competing Interests

The authors declare that he has no competing interests.

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Authors' Contributions

K.B. performed the research and wrote the paper. A.B. and K.Z. supervised the project and provided critical revisions. All authors approved the final version of the manuscript.

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