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Perovskite solar cell performance assessment

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E-mail: eva.unger@helmholtz-berlin.de, eva.unger@chemphys.lu.se and antonio.abate@helmholtz-berlin.de**Keywords:** maximum power point tracking, stability, testing protocols, current-voltage transients, hysteresis**Abstract**

Astounding progress in achieved power conversion efficiencies of solar cells based on metal-halide perovskite semiconductors has been achieved. Viable assessment of the long-term device performance is, therefore, now the most critical aspect to reliably predict device's long-term performance. Standard testing protocols to enable cross-laboratory comparison need to be established and adopted. Apart from protocols targeting the assessment of device performance and stability, procedures to investigate potential meta-stabilities in devices under different operation conditions are required to describe degradation mechanisms. This understanding will guide further optimization of materials and devices. In this perspective, we emphasize the importance of wide-spread reporting of experimental data in common databases to keep track of the state-of-the-art of perovskite solar cell performance and stability achieved.

1. Introduction

The field of metal-halide perovskite (MHP) semiconductor research has been a very dynamic one since the utilization of MHP semiconductors in solar energy conversion devices. From the start, potential intrinsic material (in)stabilities and means to omit these by material and device engineering has been a strong focus in MHP research.

Complex transient phenomena during device operation have been a significant concern and pose considerable difficulties to correctly assess the steady-state device performance from standard current-voltage measurements due to an often severe hysteresis in the early generation of devices [1–4]. This behaviour led to various reports suggesting increasingly elaborate measurement protocols, requirements for data to be provided upon publishing [5], and independent power conversion efficiency (PCE) certification.

To ensure scientific reporting supports the technological development of MHP-based solar cells, the inter-comparability of device performance and stability assessed by research laboratories world-wide is a prerequisite. A wide-spread adaptation and utilization of standard testing protocols is of utmost importance. An essential step in this regard has been the recent publication of stability testing protocols according to the International Summit on Organic PV Stability (ISOS) by a number of prominent researchers in the community [6].

Which exact measurement protocol to follow or complementary information to supply to ensure reported solar cell devices contribute to a scientific or technological advance, depends on the very nature of the work conducted. In this perspective, we outline a progression of considerations regarding the performance and stability assessment of solar energy conversion materials from 1) intrinsic material stability, 2) standardized testing of device performance, 3) degradation mechanisms derived from long-term evaluation of devices as well as 4) considerations that become important when scaling devices to larger area module technology.

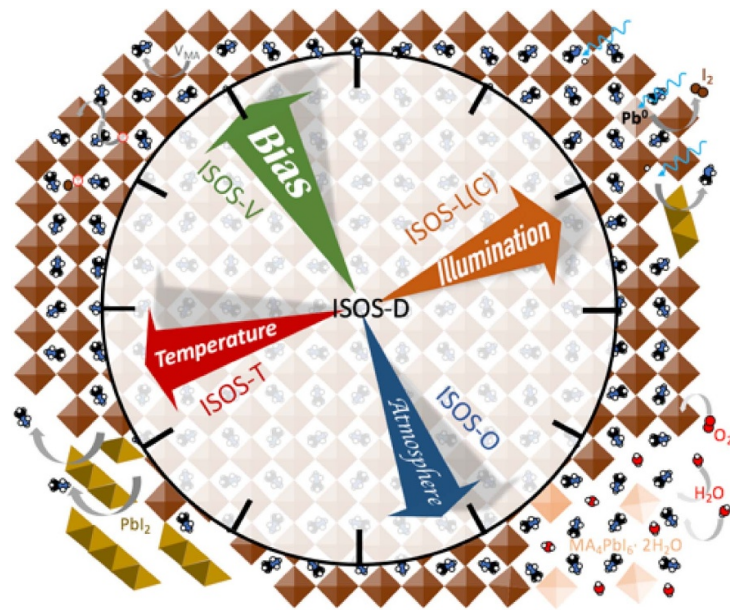


Figure 1. Schematic picture illustrating various potential degradation pathways induced by illumination, atmosphere, temperature and bias known for the archetypical methylammonium lead iodide perovskite semiconductor (MAPbI_3) and associated standard ISOS testing protocols.

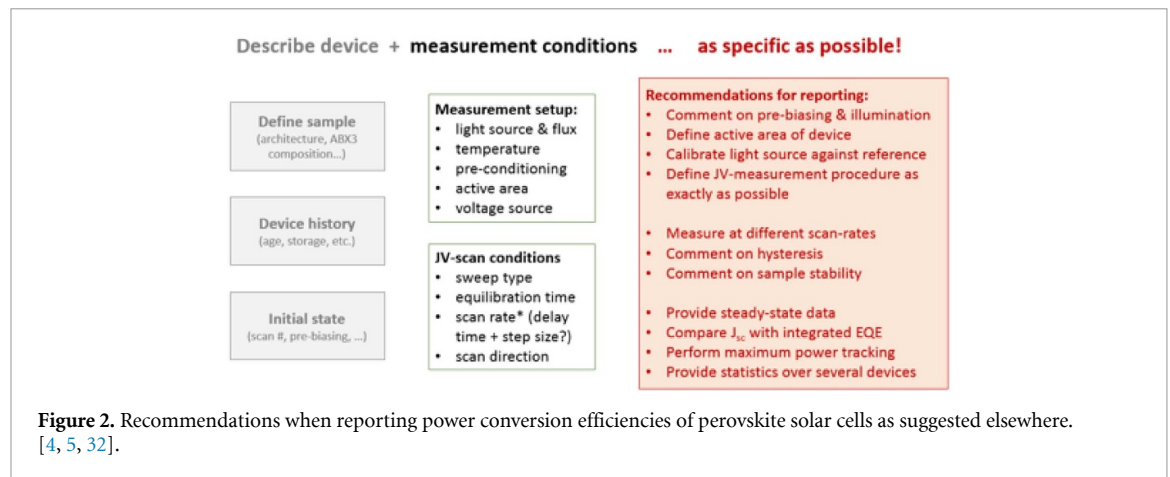
2. Intrinsic material (In)stability

In solar cell devices under operation, absorber materials can be expected to experience several stimuli that could induce reversible and irreversible changes to the materials composition. The details of material degradation mechanisms of MHP semiconductors has been summarised in detail elsewhere [7–9]. We therefore only briefly outline known degradation pathways induced by illumination, ingress of ambient species (water and oxygen from the atmosphere), temperature and applied bias of MHPs. Each of these stressors may individually or in combination lead to the degradation of an absorber in a solar cell and hence play a role when carrying out long-term stability testing according to e.g. ISOS protocols, as illustrated in figure 1 [6, 10].

Degradation in different conditions has been studied in most detail for the archetypical methylammonium lead iodide (MAPbI_3) perovskite. Senocrate *et al* [11] analyzed the thermodynamic stability of methylammonium based perovskite semiconductors under higher temperature, applied bias, illumination and in the presence of oxygen (O_2) and water (H_2O). They conclude that the Gibbs energy favours degradation under most conditions with MAPbI_3 being often the most unstable material.

Light has a somewhat ambivalent effect on MHPs as it, apart from inducing degradation, seems to be able to activate or cure certain defects, causing intermittent phenomena in the luminescence of MHP materials [12–14], which are strongly dependent on the ambient atmosphere during the experiment [15, 16], have been attributed to dynamic defects that can be in an on or off state. Other known photo-degradation mechanisms of MAPbI_3 include the structural collapse by deprotonation of the methylammonium cation [17] and photo-reduction of divalent lead (Pb^{2+}) to Pb^0 [18, 19], causing non-radiative recombination channels. Also, ionic bonds of the metal-halide lattice might be broken, generating ionic vacancies and their corresponding interstitial defects that may migrate through the material. Equally important, MAPbI_3 decomposes to PbI_2 at temperatures above 150°C [20]. Increased thermal stability of MHP materials has been achieved by cation replacement [21]. Compositional engineering has proven to be a valid strategy to engineer perovskite semiconductors with higher intrinsic stability. Still, the inherent material stability would have to sustain operating conditions for years to offer viable technological use for solar energy conversion.

Ambient species such as water and oxygen are found to affect the optoelectronic properties of MHP materials. Exposing MAPbI_3 to ambient humidity leads to the formation of hydrate phases such as $\text{MA}_4\text{PbI}_6 \cdot 2\text{H}_2\text{O}$. While the hydration is reversible, the formation of the hydrate intermediate phase negatively affects film morphology and can cause mechanical stress in the device [22]. The presence of oxygen has been found to cause peculiarities during the spectroscopic investigation of MAPbI_3 with the photoluminescence often changing dramatically [12, 23] suggesting photo-induced changes in the trap state



distribution or occupation. Superoxide O_2^- has been proposed to form charge-transfer states with $MAPbI_3$ [16, 24] and has been suggested to compensate for ionic defects in the material. Oxygen has been found to diffuse into the halide perovskite lattice, and upon illumination, $MAPbI_3$ was observed to rapidly degrade [25]. Detrimental effects on material stability due to environmental impact can be rectified on a device level by encapsulation, modification/passivation of interfaces as well as the introduction of buffer layers [26, 27].

Apart from degradation pathways due to a single stimulus, the combined effect of several stimuli needs to be considered and investigated in devices and materials exposed to operating conditions as they might accelerate or offset each other. Experimental conditions have to be carefully monitored and detailed in reports.

3. Current-voltage hysteresis indicates meta-stability

The debate around current-voltage hysteresis in the early days of MHP solar cell research highlights the importance of comprehensible and standardization of measurement routines when reporting power conversion efficiencies of perovskite solar cells [1–4]. This is in particular reflected in check-lists introduced by e.g. the Nature publishing group when solar cell device performance metrics are being reported already in 2015 [5]. Detailed guidelines on how to assess and report solar cell device performance metrics and what pitfalls to avoid have been published previously [28, 29]. Hysteresis as a phenomena and its underlying causes has been the subject of various review articles [30, 31] and book chapters [32, 33].

Due to a strong dependence of the device performance metrics on measurement conditions, scan rate and direction, device performance metrics derived from IV-measurements performed in different laboratories can be challenging to compare. Several recommendations and requirements have been proposed when publishing perovskite solar cell data, summarised in figure 2 [1–4, 32]. Even when following rigid testing and reporting procedures, there is still insecurity regarding the validity of device performance metrics as these may change dramatically due to changes in trap-state occupation or ion distribution within a device during operation or preconditioning.

Hysteresis has been linked to the displacement and accumulation of ionic species such as constituent ions of the metal-halide lattice and their associated vacancies [3]. Transient phenomena in current-voltage measurements could be either directly associated with the migration and accumulation of ions or be caused by their interaction with electronic charge carriers. Ionic defect formation and migration could reduce device performance and amplify transient capacitive phenomena over the operational lifetime of a MHP device [34]. Hysteresis and transient phenomena in the current-voltage response, in general, are hence indicators of the devices or absorber materials meta-stabilities that can negatively affect the long-term device stability.

As an example, Tress *et al* [3], investigated changes in device performance and hysteresis upon ageing at open-circuit conditions under constant 1 sun illumination comparable to ISOS-L conditions. Interestingly, hysteresis for slow scan rates was found reduced upon ageing but increased for fast scan rates. In another example [35], a strong correlation between light/bias-induced device degradation and the increase of hysteresis in mixed cation/anion perovskite solar cells was demonstrated. Exposing devices to cycles of forward and reverse biasing, fill factor and open circuit voltage were found to be reversibly affected by biasing suggesting changes in the charge carrier recombination rate due to ion redistribution. These examples illustrate that ageing may alter not only reduce device performance but also alter transient phenomena apparent in different time domains of device operation.

The transient phenomena causing hysteresis may occur on different time scales of device testing. They are, therefore, an important indication of potential meta-stabilities in the devices that may manifest as irreversible degradation already during initial testing of devices. Utilization and development of methods to analyze the transient device response in different time domains, such as impedance measurements [36, 37] or other time-resolved methods [38, 39] is required. Linking the transient decay of device performance to chemical or physical changes in the MHP material is intricate. It often requires the correlative investigation of changes in absorber materials and at interfaces in devices. Distinguishing between reversible and irreversible loss mechanisms, in particular, will enable the research community to engineer more robust materials and device stacks that are inert to these degradation pathways.

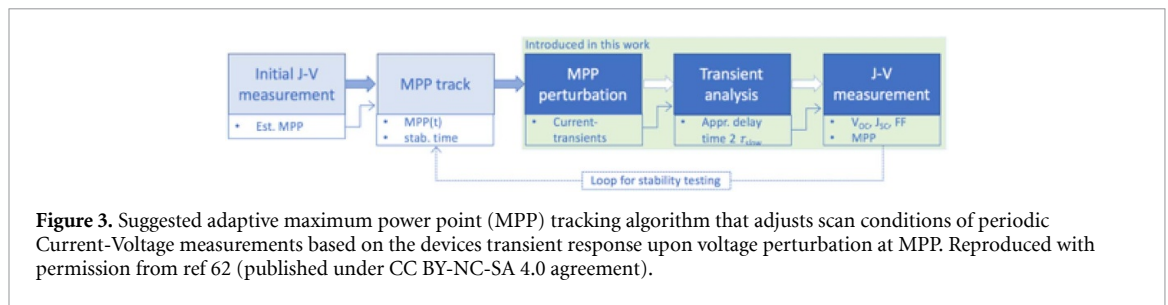
4. MPP tracking

During shorter and longer-term device performance assessment, transient effects on a slower time-scale on the order of hours or days may become apparent. MHP-devices have been shown to exhibit dynamic performance fluctuations and light/dark cycling may increase the life-span of a device due to recovery in the dark [40–42]. The origin of this phenomena is hypothesized to be due to bias-induced ion redistribution of larger organic cations [43, 44]. Recovery during dark periods suggests the light-induced formation or annihilation of deep-level trap states [14, 42]. This reversible dark/light performance fluctuation is often observed in n-i-p devices [45, 46] but not universally for all types of MHP-based solar cells suggesting that device architecture and interfaces, in particular, give rise to these transients. Changing behaviour from light-induced performance decrease to increase during an ageing experiment has been reported [46]. This behaviour highlights how dramatically the current-voltage response may change in a device under constant illumination as a function of bias and ageing conditions. It is therefore advisable to assess changes in the devices transient response and degree of hysteresis upon ageing as defects and mobile ionic species may be generated in the device upon degradation.

Transient effects make measurements of MHP-devices in actual ‘steady-state’ difficult as comprehensively summarized by Dunbar *et al* [47]. The attempt to quantify hysteresis in terms of hysteresis indices or factors is often found of limited scientific significance as these metrics may indeed just be reflecting a specific situation in a non-equilibrium condition [48]. Performing current-voltage measurements at slower and slower scan rates at some become experimentally unfeasible. Performing MPP measurements has, therefore, become a standard in the community and is often provided or required upon publishing MHP-based PCE metrics. MPP measurements are usually carried out at a fixed voltage close to the MPP derived from current-voltage measurements [40, 49, 50]. However, Performing MPP analysis based on the voltage at the MPP, V_{MPP} , determined from a single I–V measurement is not recommended since V_{MPP} can vary significantly between I–V measurements at different scan rates [51]. Preferably, MPP should be carried out using adaptive ‘perturb-and-observe’ algorithms that continuously adjust the applied potential to maximize to the total power output of the device [51–56].

MPP measurements are therefore of increasing importance and could, to some extent, replace current-voltage measures as the standard methodology to determine device performance in MHP solar cells [48, 57]. For devices such as MHP-based solar cells where ion re-equilibration in response to a change in applied potential are at play, adaptive MPP-tracking algorithms that dynamically adjust the sampling time and voltage step size will be of great value [51, 53, 54]. In this respect, Pellet *et al* [53] developed a hill-climbing algorithm to assess the MPP in hysteretic solar cell devices showing that poling devices at voltages larger than MPP can positively affect device performance. Also, Cimaroli *et al* [54], proposed a predictive MPPT algorithm that predicts the steady-state power by fitting the current response to a voltage perturbation with a biexponential function analogous to the equation.

We recently suggested an algorithm that predicts suitable settings for current-voltage measurements from the characteristic transient device response to a voltage perturbation, illustrated in figure 3 [58, 59]. The measurement principle is based on a perturb-and-observe MPP tracking algorithm, where the periodic voltage perturbation to re-assess the MPP is simultaneously used to gain insight into the dynamic current response of the device under investigation. The time constants of current transients allow the determination of a minimum delay time to carry out J–V measurements under quasi-steady-state conditions. This procedure would allow to continuously adapt the measurement conditions as devices exhibit changes in their dynamic response due to reversible and irreversible degradation during operation. Such an adaptive algorithm can be easily implemented as an automated measurement routine.



5. Long-term performance and higher technology readiness level (TRL)

Once the general feasibility of solar energy conversion by novel materials and devices is demonstrated with high PCE, the long-term energy conversion becomes a more significant metric.

MPP measurements can, in principle, be expanded to arbitrary long duration to assess the long-term performance losses under maximum power operating conditions. To account for fast capacitive phenomena, MPP tracking for about 5 min is usually found to be sufficient to confirm device performance metrics derived from current-voltage measurements. Once a certain state-of-the-art is reached, which for MHP-based devices would be a PCE of >20%, assessing the longer-term stability of materials and devices are often required. While it is usually feasible to supply medium-term stability data of single devices on the order of 12–24 h. To track devices performance for longer durations on the order of 1000 h, specialized testing equipment is required and few research groups hence provide systematic stability data and even fewer can carry out studies including statistics from larger batch sizes of devices studied for a single parameter [26, 27].

In addition to the measurement algorithm, discussed in the previous section, the metric or analytical procedure to capture device stability in a single metric is the target for the comparison. This target is, however, not very straight forward. The T_{80} value can be a misleading figure of merit for devices with substantial transient phenomena during the initial phase of device operation [46, 60, 61]. This has led to proposing the use of an integral lifetime energy yield $LEY = \int_0^t PCE(t) dt$, as a more reliable figure of merit to reflect the device performance over time [46]. With respect to specifying measurement conditions, defined measurement protocols such as ISOS [6, 10] need to be employed as the device degradation strongly depends on the devices operating conditions [62]. This will enable inter-laboratory comparability and facilitate establishing degradation mechanisms of materials and devices alike. These should be carried out already on lower TRLs based on laboratory test devices.

Once reliable encapsulation strategies for devices are in place, the simulation or measurement of devices under ‘real conditions’ becomes very interesting [63]. On this level, the robustness of encapsulation materials and mechanical stresses arising in device during temperature cycling become of interest [64, 65].

Especially for device technology entering higher TRLs, testing devices and modules according to international standards such as the IEC 61215 standard [66, 67] becomes relevant [68]. Few studies have employed this standard to test the efficacy of devices and encapsulation strategies to date [64, 69, 70]. In modules, degradation due to reverse biasing may become a new pathway of device failure [71]. Recently, the potential leakage of lead from MHP-based modules and role of encapsulation materials to minimize lead entering the environment after mechanical damage of modules has been investigated [72]. Now that MHP-based device technology is being developed on higher TRL levels for the first generation of efficient MHP-semiconductors, studies assessing the overall deployability of these devices are needed. Data reflecting the real operational risk of heavy-metal leakage would provide essential guidelines for further device development.

6. Measurement standards & OpenScience

Considering all the above, standardized performance and stability assessment protocols are of particularly crucial importance in evaluating the short-term as well as the long-term performance of perovskite solar cell devices. This goal is a prerequisite to assess the technological potential of perovskite solar cell devices at which point device prototype evaluation according to IEC standards will become relevant [66, 67].

Device and material assessment on lower TRLs require the analysis of device failure mechanisms as a function of degrading stimuli applied. In this regard, the recently published consensus paper by more than 50 leading scientists in perovskite photovoltaics on standardized ISOS stability testing protocols is an important step to establish defined stability assessment protocols that allow inter-comparison between

research laboratories that will provide a larger dataset for the identification of degradation pathways of perovskite solar cells [6, 10].

For performance assessment by current-voltage measurements, adherence to standardized procedures is required. There are numerous recommendations available on how to correctly assess and confirm experimental performance metrics of solar cell devices.

When publishing a new performance record for a particular semiconductor material, device architecture, module or tandem device incorporating MHP-based materials, independent certification by an accredited laboratory can be an essential step to establish a new reference point representing the state-of-the-art of this solar cell technology. Sending devices for independent certification takes time, often money and may risk device degradation during shipment and handling elsewhere. To some extent, independent certification ensures that devices are sufficiently stable to be tested elsewhere. This is an important step in validating a technology and enabling complementary measurements in other labs.

OpenScience reporting and publishing requirements will become a desired necessity as much insecurity and issues in comparability will be omitted when (step 1) adhering to relatively rigid testing protocols for J-V measurements accepted and adopted by the majority of the research community including (step 2) facilitating measures in different research laboratories by making code available for software-controlled measurement procedures. In the future, we foresee significant progress in science if extensive datasets are gathered according to standardized testing protocols and made available in Open Databases. This approach will enable a self-updating representation of the state-of-the-art of perovskite solar cell performance and stability. Creating a more significant fraction of datasets available to scientific scrutiny will facilitate advances in sustainable power conversion technology driven by scientific insight.

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