The challenge of designing accelerated indoor tests to predict the outdoor lifetime of perovskite solar cells

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The challenge of designing accelerated indoor tests to predict the outdoor lifetime of perovskite solar cells

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Abstract

Over the past decade, perovskite solar cells have travelled an amazing way towards high efficiency. However, a major roadblock remaining is the operational stability, while achieving technological maturity and proving real-world stability is crucial to gain trust among investors. In that sense, it is of high interest to be able to predict the operational lifetime, which needs to be in the range of years or decades, within an experimentally reasonable timeframe. Yet, peculiarities of perovskite solar cells' ageing behaviour lead to severe difficulties in translating the results of indoor tests to their outdoor counterpart. In particular, transient processes cause diverse results among different ageing tests.

Here, for the first time, we show a complete set of constant illumination indoor testing, cycled illumination indoor testing and real-world outdoor testing on equal in-house devices. Exemplarily, we compare two different types of perovskite solar cells, in which only the hole-transport layer is varied. Despite this small change, the devices show distinctly different transient behaviour. In either case, the commonly used constant illumination experiments fail to predict the outdoor behaviour of the cell. Yet, we observe a good correlation between the cycled illumination test and the outdoor behaviour of one of the two solar cells, while this is not the case for the other system. This result highlights the urge for further research on how to perform meaningful accelerated indoor tests to predict the outdoor lifetime of perovskite solar cells.

Introduction

Perovskite solar cells (PSCs) are a rapidly developing photovoltaic technology[1]. The next step on the path towards commercialisation is to surmount the challenge of stability under outdoor operation conditions. Despite significant improvement of PSC stability with respect to light, heat, and humidity achieved over recent years[2,3], this class of solar cells' stability under outdoor operation remains almost unexplored. This is one of the first studies reporting on maximum-power-point-tracked outdoor installed PSCs samples[4]. Obviously, understanding real-world device operation and stability is an essential element in technology maturing and fosters trust in the new technology among investors. That trust in the reliability of a technology is especially important in photovoltaics, as expected service time is in the range of 25-30 years. The required investment is almost completely required initially and commercial viability depends crucially on minimal and predictable technical degradation of the photovoltaic installation. In order to guide the development of accelerated ageing tests in the lab, we present first insights into the challenges of translating the results of indoor tests to the real-world performance of perovskite solar cells.

Over the past years, it took significant efforts to develop reliable and reproducible laboratory measurements for testing PSCs' efficiency. This difficulty was mainly due to ionic movements[5,6] and other slow transient processes[7] affecting perovskite solar cells. Thus, PSCs are not compatible with the classical methods used to estimate current-voltage characteristics (JV). Broad discussion in the research community has resulted in widely recognised common practices for efficiency estimation under standard test conditions (STC)[8,9]. The most obvious conclusion was introducing maximum power point tracking (MPPT) to validate the value extracted from JV scans.

More recently, the influence of transient behaviour in PSCs was found relevant in studying the long-term stability of devices [10,11]. The origin of that behaviour is the coexistence of several dynamics with characteristic times spanning from timescales of seconds to hours[12]. Such slow dynamics are relevant in the day-night cycling of devices in operational condition and need to be considered to estimate PSCs' energy yield over the device's lifespan. Although a consensus on the particular ageing tests to assess PSCs' stability has been agreed on[13], there is no clear strategy to predict the PSCs' *outdoor* lifetime from accelerated indoor ageing. Despite the scarce experience in this area, available data already suggests that innovative ideas are needed to overcome the challenges[11,14]. The main challenges include a diverse diurnal behaviour of perovskite cells caused by slow transient processes that affect the performance over the day-night or degradation-recovery cycle. Those transients, whose nature depends on the type of solar cell investigated, make the extraction of cell parameters (*e.g.*

temperature coefficients) from outdoor data unreliable and complicate the investigation on the relation between outdoor and standard indoor tests.

This work analyses available literature and in-house data on PSCs' outdoor and indoor ageing behaviour, outlines challenges in predicting operational lifetime through accelerated ageing tests and suggests strategies to overcome those.

Outdoor Testing

To achieve the prediction of a realistic outdoor lifetime of PSCs with an accelerated indoor test, it is essential to understand the temporary changes in device power output due to weather conditions and the state of the device. This section will discuss the shortcomings of existing PSCs' outdoor data thus far, showcase the difficulty to extract valid device parameters from outdoor tests, and provide examples of different transient processes in PSCs that affect their energy yield and lifetime.

Despite the extensive research activity in the field of perovskite solar cells, only few papers so far reported the cells' behaviour under outdoor conditions, amongst those only one under MPP-tracking. Table S1 provides a summary of the publications known to the authors. The majority of these papers focuses on PSCs' stability and utilises outdoor conditions as a realistic combination of stress factors. According to the ISOS-protocols[13,15], outdoor-related ageing experiments are classified into three groups. While ageing occurs under outdoor conditions in all protocols, the characterising measurements and the electronic load during ageing are varied. According to ISOS-O-1 periodic JV-measurements are performed under a solar simulator indoor, while ISOS-O-2 requires the periodic acquisition of JV-curves under the natural solar light outdoors. Whereas in those protocols, the electronic load during ageing outdoors can be MPP *or* open circuit, ISOS-O-3 demands for continuous maximum power point tracking and periodic JV-scans in STC. Almost all reports so far rely on the two former protocols, and mostly the devices were at open circuit during the ageing. However, although not specific to PSCs[16], the electric load condition is critical particularly in perovskite-based devices, mainly due to their sensitivity to ion redistribution[17]. The operational load has significant implications on the degradation mechanisms and measured device lifetime[18]. PSCs' degradation is often (but not always[19]) fastest at open circuit[18]. Also, in any real application, the solar cell will be operated at MPP. The strong dependence of PSCs' ageing behaviour on the electronic load and MPPT being the only realistic load condition highlights the importance of conducting ageing experiments under MPPT.

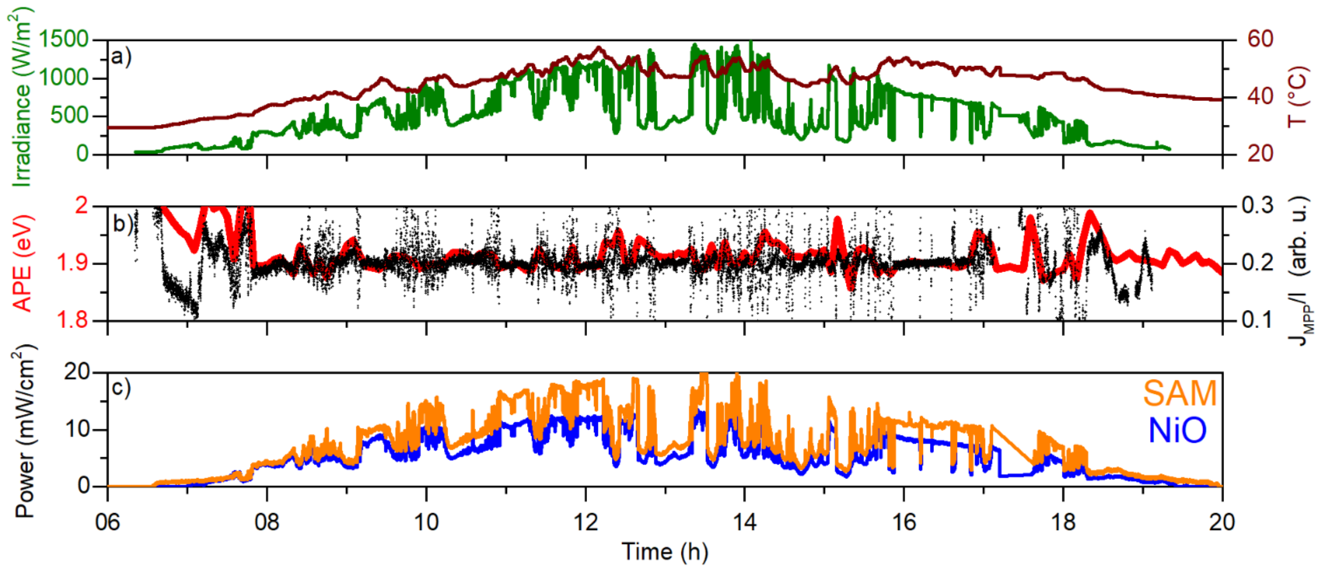

Figure 1: Impact of real-world stresses on the PSC's power output: One day of outdoor exposure of PSCs with organic self-assembled monolayers (SAM) and NiO hole transport layers. The respective power output c) measured via MPP tracking closely follows the measured irradiance a). The average photon energy (APE) in b), is calculated from spectral data measured in the plane of samples and plotted next to the ratio of J_{mpp} to irradiance (black dots in b)) to highlight the effect of the sun spectrum.

Figure 1 shows an example of MPP output from two different p-i-n PSCs during one day of outdoor testing together with measured irradiance and temperature during that day. The examined cells are comprised of the following layer stack: ITO/HTL/$Cs_5(MA_{15}\,FA_{85})_{95}Pb(I_{85}Br_{15})_3/C_{60}/SnO_2$/Cu; the hole-transport-layer (HTL) was varied. As one option for the HTL, NiO was used, which is the "standard" for stable p-i-n devices[20], while as other HTL the newly developed self-assembled-monolayer (SAM) "MeO-2PACz"[21] ([2-(3,6-dimethoxy-9H-carbazol-9-yl)ethyl]phosphonic acid) was used. A variant of this molecule, the "MeO-4PACz", recently gained attention since it enabled a record perovskite-silicon monolithic tandem solar cell[1,22].

As with other PV technologies, PSCs' power output depends on irradiance, temperature, solar light spectrum, and incidence angle. As can be seen from Figure 1 a) and c), the power output of the solar cells resembles the measured irradiance, which is due to the linear dependence between irradiance and MPP current density (J_{MPP}), see Figure S3. Yet, the J_{MPP}-to-irradiance ratio is not a straight line as shown in Figure 1b. Some deviations from this linearity are caused by changes in the incident sunlight spectrum. They coincide with changes in the average photon energy (APE) of the incident spectrum, which is a technology-agnostic figure of merit of the spectrum (see Eq. S1 for the definition)[23,24]. In essence, a blueshift (higher average photon energy) in the incident spectrum leads to a marginally improved device current, while a redshift (lower energy) leads to a marginal decrease.

With outdoor data available, a logical next step to take -also for investors- would be energy yield predictions and analyses [25–27]. However, we advise caution here, since the extraction of parameters from outdoor data might lead to a misinterpretation in case of PSCs. For example, E. Velilla *et al.* found temperature coefficients insignificant for PSCs [26]. Stoichkov *et al.* even observed PSCs' temperature coefficients being positive in some cases when derived from outdoor data[25], meaning that device efficiency improves with temperature. While a positive temperature coefficient may correlate to annealing effects or ion redistribution, it might also be entirely misleading due to unaccounted transient behaviour overlapping with temperature effects in real-world conditions. Table S3 summarises reported temperature coefficients obtained from either indoor or outdoor data. When measured indoor, temperature coefficients of PSCs are negative like in all other PV technologies, lying in the range of approximately -0.1 to -0.3 %/K. An exception poses devices with Spiro-OMeTAD, which show a non-linear dependence due to a peculiarity of this hole transporting material [28,29].

Since reports of PSC temperature coefficients are scarce at the moment, in Figure 2, we compare the temperature coefficients at different light intensities derived from indoor and outdoor measurements on the same set of samples. The two different hole-transport layers are used in an otherwise identical stack. The devices show temperature coefficients from app. -0.1 to -0.3 %/K when measured in a controlled indoor environment after saturating the light-soaking effect (see SI for details), which is in the expected range. However, we observe significantly different values from outdoor data on the same type of devices, including some positive coefficients (see SI for details of calculation). Positive correlation in outdoor power with temperature was also reported to GaAs solar cells and explained by temperature-correlated spectral changes[30], which we believe cannot explain results reported here. Instead, we believe that the discrepancy shown in Fig. 2 arises due to the presence of reversible processes that affect the PSC's daily behaviour and hence prohibit straightforward data interpretation. Indeed, the hottest part of the day is typically midday, meaning that the natural periodic increase in device temperature overlaps with reversible periodic transient processes in PSCs, affecting calculated values. This example showcases that caution must be taken when extracting PSC parameters from outdoor data.

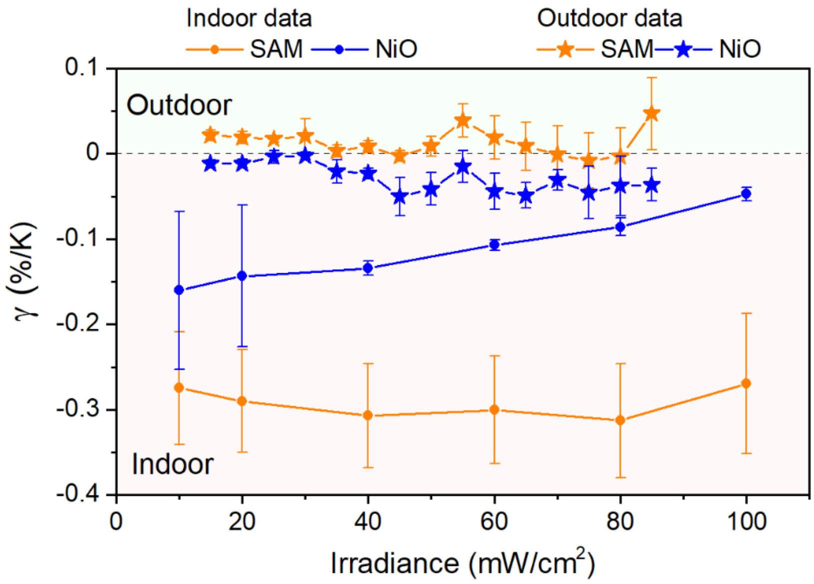

Figure 2: Care needs to be taken when extracting device parameters from outdoor measurements: Temperature coefficients of two types of PSCs with different HTMs at different light intensities, calculated from indoor and outdoor measurements.

Transient effects not only complicate the extraction of device parameters, but also influence the diurnal behaviour in outdoor conditions. Two different reversible transient patterns have been reported for PSCs[10]. In one case, cells are losing efficiency upon operation and recover during the following dark period. The opposite is also known: some perovskite cells improve upon light soaking and lose efficiency when put into the dark. It was also observed that the time under illumination required to reach peak performance might increase with consecutive cycles. This effect is called fatigue. In this paper, we refer to *fatigue* as any change in shape or absolute height of MPP-efficiency over time curves with increasing number of cycles.[31,32] Transient processes in PSCs can occur on a timescale of hours (sometimes even hundreds of hours[33]) under illumination[34] or in the dark[35]. The exact type of PCE evolution varies depending on the device stack and degradation stage[12].

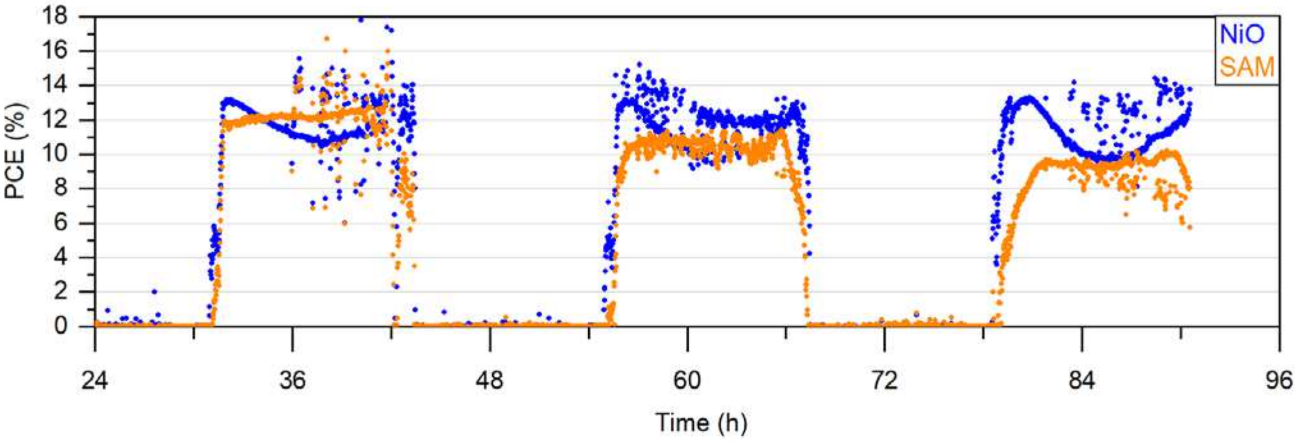

Figure 3: Exemplary PCE of two types of PSCs with different HTMs during three consecutive days of outdoor MPP-tracking

As an example, Figure 3 shows the power conversion efficiency of two different stacks of p-i-n PSCs under MPPT in outdoor conditions during three consecutive days. While the only difference between stacks is the hole transport layer, these two types of cells show dramatically different day-night behaviour patterns. PSCs with NiO perform best in the morning, slowly degrade over the day and recover to almost initial values overnight, while already starting to recover in the evening hours. Relatively high series resistances contribute to this behaviour, possibly explaining a better performance at low irradiance in the evening (see Figure S4). However, since evening values are still noticeably below morning ones, we assume the impact of reversible degradation. On the contrary, PSCs with SAM require several hours (strongly depending on the day of observation) to reach their peak efficiency.

It is not always easy to judge such transient behaviour from indoor experiments, partly due to frequent practice of removing the initial "stabilisation" phase from the reported PSC ageing curves. We strongly recommend providing full datasets, because under the natural day-night cycle in outdoor conditions, initial transient processes might have a significant contribution to the observed behaviour. Although not necessarily detrimental for the long-term device stability in a constant light experiment, such transients might significantly contribute to PSCs energy yield in outdoor tests. These transients have to be considered for optimising the devices for outdoor applications. We encourage the community to report more information on the devices' light-soaking behaviour and recovery even for standard constant illumination experiments, like the time-to-maximum, time-to-saturation of recovery and the extent of recovery (see Figure S5 for schematics).

The presence of reversible processes makes the commonly used constant illumination tests a suboptimal proxy for device stability and complicate the comparison between indoor and outdoor ageing tests in PSCs. Transients changing with each cycle obviously cannot be present in a constant illumination test. This challenge will be discussed in detail in the next section.

Comparing Indoor and Outdoor

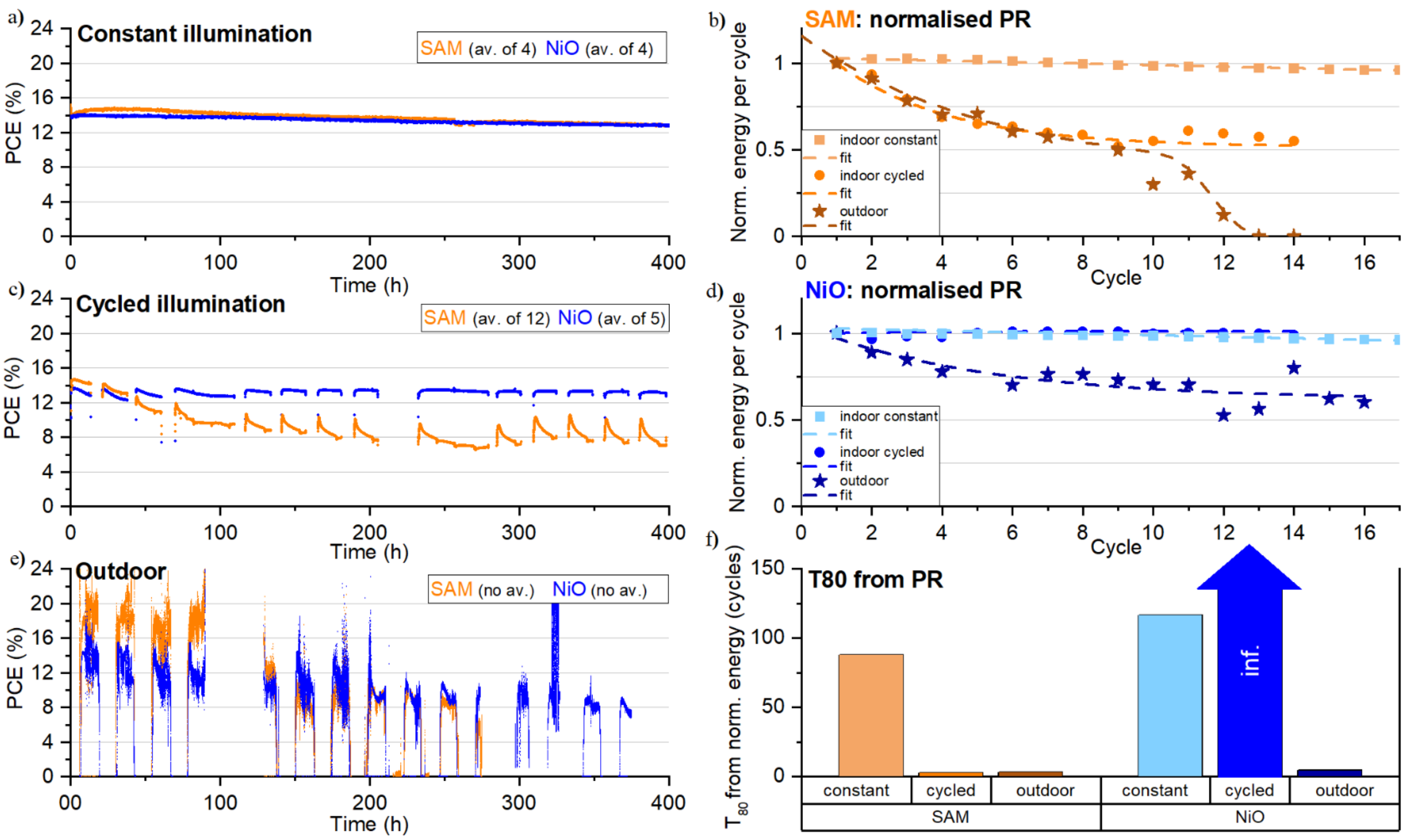

Figure 4: Comparison of indoor and outdoor stability for two solar cells using a different HTL. a) Indoor experiment with constant illumination. c) Indoor experiment with cycled illumination. Both indoor experiments are conducted at 25 °C and in nitrogen. e) Outdoor experiment, conducted in Berlin, July-August 2020. Values not averaged for better display; averaged performance ratio over 5 and 6 cells as well as another outdoor run in Figure S6. All experiments a) c) e) are performed under continuous MPP-tracking. b),d): Integrated power output divided by integrated irradiance (performance ratio), integrated over 12 h, one cycle or one day, then normalised to the first day. f) T_{80} calculated from b) and d).

As a first step towards predicting outdoor lifetime with indoor testing, we present a comparative study with indoor and outdoor data. Three different ageing tests, indoor and outdoor, have been performed (see Table 1).

In our study, both types of solar cells were tested at first under constant ageing conditions indoors. These were in brief: 100 mW/cm^2 continuous illumination by a solar simulator (Figure S1 displays the spectrum), MPP-tracking, 25 °C actively controlled sample temperature, measurements under nitrogen atmosphere. The test corresponds to the protocol ISOS-L1-I, and the results are shown in Figure 4a (averages of 4 cells each). Secondly, a cycled indoor experiment was performed on another set of samples. It was conducted under the same conditions, but the illumination was switched on for periods between 13 h and 38 h. Then, cells were put into the dark and disconnected for periods between 8 h and 25 h (see Figure 4c, averages of 5 and 12 cells). The test corresponds to ISOS-LC-1-I. Finally, another set of samples were encapsulated between two glasses using acrylic glue and exposed outdoors with continuous MPP tracking (Berlin, July-August 2020) according to the ageing protocol ISOS-O-2 (see Figure 4e, averages of 5 and 6 cells). Note that there is a data gap from 4^{th} day evening to 6^{th} day morning. Weather conditions (irradiance, ambient and cell temperature, relative humidity) were simultaneously recorded and are shown in Figure S7.

Table 1: Conditions of the performed ageing tests.

	Test 1 "Constant illumination"	**Test 2 "Cycled illumination"**	**Test 3 "Outdoor"**
light source	simulated light, 100 mW/cm^2	simulated light, 100 mW/cm^2	solar light
illumination	constant	cycled	varying
temperature	25 °C actively controlled	25 °C actively controlled	varying
electronic load	MPP-tracking	MPP-tracking	MPP-tracking
test atmosphere	N_2 flow	N_2 flow	air (encapsulated)
encapsulation	none	none	glass to glass, acrylic glue, N_2
system	open	open	closed
ISOS protocol	ISOS-L1I	ISOS-LC-1I	ISOS-O-2
Figure	4a	4c	4e

When analysing the constant indoor test (Figure 4a), it seems that the overall stability is very similar. However, the tracks' slope is different, and the curves are crossing each other after ~420 hours of testing (see the full test in Figure S8). Consequently, NiO could be considered more stable in this test with a T_{80} (i.e. the time it takes for PCE to decrease 80% of initial value) of 835 h, while SAM's T_{80} equals 580 h.

The behaviour changes significantly when the illumination is cycled (Figure 4c). Within each illumination period, for both stacks, the efficiency rises very fast at first and appears almost like a vertical line at the beginning of each cycle; compare to Figure S9, where one cycle is displayed in detail. This rise takes around 1 h and does not originate from the algorithm finding the MPP since that happens within a timescale of minutes. For NiO, this initial rise in efficiency is faster and less pronounced than for SAM. After reaching the maximum value, PCE starts to drop over time until the illumination phase is over in both stacks. For SAM, this decay phase is of exponential shape, while for NiO it is less pronounced and appears to be linear. With an increasing number of cycles, the shape of the curves changes for both systems. As discussed before, this change in transient behaviour with a growing number of ageing cycles is called fatigue[31]. For cells utilising SAM, the decay becomes steeper with increasing number of cycles.

Additionally, the maximum value reached drops dramatically during the first 5 days and then saturates until day 10 for SAM. For NiO, however, the curves decay only during the first 5 days and recover to nearly the initial value during the dark phase. After 5 days, the efficiency stays mainly constant after the initial rise of each cycle. The tracks of the outdoor experiment (Figure 4e) have been discussed before. It can be highlighted here that the two stacks' transient behaviour is substantially different and seems to interplay with the changing outdoor conditions.

To compare the data, we computed the so-called performance ratio (PR) for all three ageing experiments with equation (1).

$$performance\ ratio = \frac{\int_{t_1}^{t_2} P_{mpp}\, dt}{PCE \cdot \int_{t_1}^{t_2} irradiance\, dt} \qquad \text{Equation (1)}$$

Here, the power output is integrated over a given time interval and divided by the integrated irradiance multiplied with the initial PCE measured at standard test conditions.[36] The PR therefore reflects the efficiency over the course of a full day. We then normalised the PR to the energy output of the first day.

In real operation and outdoor experiments, energy can naturally be harvested only during the daytime, and the effective illumination time depends on the location and the time of the year. Consequently, the integration was performed over 24 h for the outdoor experiment. The cycled indoor illumination period was varied between 13 and 38h; here, the performance ratio was integrated over a full illumination cycle. For the ageing experiment with constant illumination, there is no cycle. We artificially introduced integration over equal periods of 12 h in order to have a comparable figure of merit. Figure 4b shows the normalised PR per cycle of SAM-cells for all three experimental conditions; Figure 4d shows the same for NiO. The plot enables us to compare the different datasets elegantly.

Solar cells utilising SAM that are tested under constant conditions behave quite differently when light-cycled in the same conditions. In the continuous light experiment, the normalised energy per cycle shows a linear decay, while in the light-cycled experiment the decay somewhat resembles an exponential shape (compare Figure S10). Since the only difference between those two indoor experiments is cycling the light, the different behaviour likely originates in recovery effects[5,7,12]. It seems that pausing the illumination and putting the cells to open-circuit condition harms the device more than constant illumination and MPPT conditions. It appears counterintuitive that giving the cells "a rest" hurts them more than applying constant stress.

Interestingly, the indoor cycled PR for SAM-cells does match quite well with the outdoor experiment's ones until day 10. Then the performance drops rapidly over the next days until total failure on day 13. We assume that this is due to the breakdown of the encapsulation: As soon as water or oxygen have passed the encapsulation, the degradation rate is strongly enhanced and also leads to visual changes (see Figure S11) in the device due to perovskite decomposition[37]. Since passing the encapsulation is likely a diffusion process (see Figure S12), an error function can be used to model the behaviour. Hence, it instantly boosts the degradation rate, explaining the rapid drop of PR after day 10[38]. Despite this deviation, we were successful in reproducing the outdoor behaviour in a controlled indoor experiment in terms of trend and behaviour in this case. Still, it is evident that the two decay curves' exponential constants are different, and a lifetime prediction from the cycled indoor experiment would be erroneous. Also, it should be noted that the cycled indoor experiment is not yet accelerated since the actual test time was the same in both cases.

Despite these promising results for solar cells utilising SAM, the indoor cycled experiment's PR curves and the outdoor experiment’s ones significantly deviate from each other for NiO. The outdoor curve again shows exponential decay, but the indoor cycled curve shows a linear behaviour without noticeable decay. The indoor cycled curve matches much better with the constant indoor one, which also shows linear behaviour but with a negative slope. This means that for NiO, the cycled cells show higher stability than in the experiment with constant illumination, which is the exact opposite behaviour to SAM.

The transient and fatigue behaviour appears to be fundamentally different for the two systems: It appears that with NiO, switching the light on-off is helping the devices to stay stable, while for SAM, the light switching is diminishing the cells' lifetime. The constant light experiment does not match the outdoor behaviour in either system. These main findings are also represented in Figure 4f, which displays T_{80} calculated from the normalised PR. For SAM, the outdoor behaviour was at least depicted properly by the indoor cycled experiment. However, an outdoor lifetime prediction from the indoor cycled experiment would be wrong for NiO. For this stack, we could not depict the outdoor behaviour with the indoor tests.

Without further experiments, it is difficult to tell why the stacks' behaviour is diverse, even if the only difference is the HTL. Possible reasons might comprise ion migration and possibly passivation effects, chemical reactions at the interface or the degradation of the HTL itself.

In conclusion, we found that outdoor behaviour could be reproduced with an indoor cycled experiment for cells utilising SAM. In contrast, for cells with NiO, outdoor behaviour could not be reproduced indoors. The difference is due to the diverse fatigue behaviour of the two systems.

Strategies for accelerated indoor testing

Our endeavour is to conclude from an accelerated indoor test to perovskite solar cells' outdoor behaviour. The transient and fatigue behaviour of perovskite solar cells represent the main challenge. To tackle that challenge, there are three strategies: (1) to develop devices with minimal transient and fatigue behaviour, (2) to establish advanced indoor tests that are able to closely emulate outdoor degradation patterns and (3) to perform several indoor tests that deconvolve the degradation mechanisms present under the real-world combination of stresses.

Minimizing the transient behaviour as in (1) is one target of current PSCs research. Yet, so far, this has not been reached and consequently the community needs to treat these transients as part of device characteristics, report them, and understand the underlying mechanisms. It might still be possible to predict outdoor behaviour even from a constant load test. However, a prerequisite for this would be that the initial transient carries all the information needed.

The second strategy (2) is to develop suitable indoor tests to emulate real operational conditions better and eventually accelerate the ageing process in a multi-stress approach. Tress et al. follow that approach and developed a testing machine that is fed with weather data (temperature and irradiance) and reproduces these in a nitrogen atmosphere[28]. Recently, also Song et al. highlighted the need for advanced indoor testing procedures[11]. Yet, such machines are costly, complex, and require a high degree of development and will not be available in many labs. Additionally, the technical complexity will make it challenging to build testing machines with sufficient capacity in order to achieve high-throughput testing and collect significant statistics. However, it might be an option if these machines become available commercially.

In the third approach (3), researchers would perform different ageing tests that focus on one specific stress factor at a time. Consequently, the real-world degradation is split into a set of degradation mechanisms, each of which can be reproduced in the lab with a different ageing test. Then it might be possible to increase each stress to accelerate the test and obtain individual acceleration factors. By collecting these individual acceleration factors, it could be possible to deduce a global acceleration factor that predicts the cells' real outdoor lifetime. This approach appears appealing, even if many tests need to be conducted carefully. One major drawback of this approach is unavoidable: If stresses trigger specific degradation paths only when combined, the tests won't depict real operation anymore.

In any case, it must be strictly avoided that the acceleration triggers additional or unnatural degradation mechanisms. Additionally, we urge researchers to report significant statistics on any ageing test performed. The common practice of reporting single-pixel ageing tracks must be seen as a starting point to tackle stability. Still, it can lead to misinterpretation and over-or underestimation of lifetimes of particular stacks.

Conclusion

Being able to predict the outdoor lifetime is crucial to speeding up the commercialisation of PSCs. Accelerated indoor tests, as standard (and standardized) as for other solar cell technologies, are obviously needed. Yet, those standards are not applicable to PSCs due to their transient behaviour that is not accounted for in constant illumination testing. We were able to show in this contribution, that indoor cycled illumination tests do represent outdoor behaviour well over a range of time for a SAM hole transporting layer PSC. However, the same test does not have predictive power for PSCs with a NiO hole transporting layer instead, indicating that simple cycling of the light is only a part of the solution in bridging indoor and outdoor ageing experiments.

Developing devices with minimal transient and fatigue behaviour is one goal of current PSCs research and would enable researchers to use existing lifetime tests. However, the pace of development may slow down commercialization. Finding indoor tests that emulate outdoor degradation patterns is necessary to come up with an adequately accurate lifetime estimate. This goes hand in hand with attempting to unravel the degradation mechanisms present under the real-world combination of stresses.

We point out the urge in finding appropriate and accelerating tests with immense care. Those tests need to be validated for their applicability to each specific PSC composition. More ageing data have to be reported fully and with statistical relevance, more outdoor data acquisition is needed.

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