Baselines for lifetime of organic solar cells

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Highlights

- Baselines for lifetime of organic solar cells tested under different ageing conditions are presented
- A list of devices with exceptional intrinsic stability is provided
- Lifetime progress diagram with best lifetime is shown

Abstract

To this date there are no reliable methods for qualifying and guaranteeing the durability of a product made from organic photovoltaics (OPVs) or other similar emerging technologies, such as dye sensitized and perovskites solar cells. The issue however has to be urgently resolved in [illegible] a part of a larger effort of developing a worldwide database of lifetimes that can help establishing reference baselines of stability performance for OPVs and other emerging PV technologies that can then be utilized for determining and predicting the lifetime of the future products. The study constitutes scanning of literature articles related to stability data of OPVs, reported until mid-2015 and collecting the reported data into a common database. A generic lifetime marker is utilized for rating the stability of various reported devices. The collected data is combined with the earlier developed and reported database, which was based on articles reported until mid-2013. The extended database is then utilized for establishing the baselines of lifetime for OPVs tested under different conditions. The work also provides the recent progress in intrinsic stability of OPVs with different architectures, as well as presents the updated diagram of the reported record lifetimes of OPVs. The presented work is another step forward towards the development of a lifetime prediction tool for emerging PV technologies.

1. Introduction

There exists a set of international standards (typically published by IEC and ASTM standards organizations) in the photovoltaic (PV) world that target specific testing and qualification methods for PV based products and enable the possibility for guaranteeing the performance of these products in the end use environment. These standards are typically suitable for silicon based and other inorganic PV technologies. Meanwhile, rapidly developing emerging PV technologies, such as organic photovoltaics (OPV), dye sensitized solar cells (DSSC), perovskite solar cells (PVSK) and others alike still lack standard testing methodologies that would allow reliably predicting their performance in the end use environment. The reason partly comes from the fact that the emerging PVs considerably differ in architecture from their inorganic counterparts [1] and due to their increased sensitivity towards the testing environments [2–5] the common testing standards are not suitable for these technologies [6]. In addition, standards are requirements and recommendations that are created by bringing together the best practices and many experiences of various expert groups in the field, and due to the relatively young age of the emerging technologies and lack of controllable testing procedures there has not been generated sufficient amount of reliable data so far that could lay the basis for development of standards.

These challenges however have received significant attention in the recent years especially in the field of OPVs. In particular, at the sequence of International Summits on Organic solar cell Stability (ISOS) reliable testing of OPVs was thoroughly addressed and in 2011 recommendations were published based on the consensus of a large number of renowned research groups in the field, that outlined recommendations for reliable stability testing of organic solar cell [7]. The guidelines set certain criteria on the test conditions and therefore allowed reproducibly recording the ageing of the samples under specific controllable conditions in both indoor and outdoor testing environments. While this very much helped in reducing the spread in the testing procedures among the different groups and improving the reproducibility of the reported device lifetimes [8], the question still remained, how to develop a methodology that would allow predicting and thus guaranteeing the lifetime of a product in end use environment based on accelerated testing. Significant efforts are put today towards resolving this and in particular, recently DTU group has demonstrated an approach based on statistical analyses, where a large set of variety of OPV samples were tested under different ISOS tests and the average lifetime of the samples under each test condition was determined [9,10]. The values were then used to calculate the ratio between the accelerated and real outdoor tests, which could potentially be utilized for predicting device performance. However, despite the relatively large data sets the studies were limited to only a few architectural variations and while they well demonstrated the concept, the established values could not be regarded as sufficiently generic for application beyond the reported studies.

The works however continued and recently a manuscript was published by the same group, where the same statistical approach was utilized for analysing the entire literature related to stability of OPVs [11]. In the study, the authors collected analysed all the articles reported until March 2013 discussing stability studies of OPVs (total of 2500 article). A generic lifetime marker was developed that allowed gauging and intercomparing the stability of the different OPV devices reported in these articles. The lifetime of the samples was categorised depending on device type and architecture and depending on the test conditions, which helped better understanding and elucidating the typical bottlenecks for the device stability. The study additionally helped establishing averages for the lifetimes of OPVs tested under different test conditions. However, due to the limited amount of data for certain test conditions (especially for outdoor data) some averages lacked statistical significance and thus, could not be regarded as reliable baselines for device lifetime. The initiative therefore continued with the purpose of further enriching the lifetime database with both literature reported and experimental data and converting the database into a generic hub of baselines for the lifetime of OPVs and other emerging PV technologies alike and utilizing the data for establishing the prediction tool.

This work, as a complementary to the aforementioned earlier reported study, presents the results of the follow up literature analyses for additional period starting from March-2013 until

March-2015. The data analysis provides more solidified distributions of the lifetimes and allows drawing conclusions on the baselines for the OPV lifetimes tested under specific conditions. An updated version of the lifetime progress diagram is presented as well.

2. Methodology

2.1 Literature data

The data collection procedure is explained in detail elsewhere [11]. Briefly the articles were identified using the search engine ScienceDirect and exploring expressions based on different combination of words such as polymer, plastic, organic, solar cells, photovoltaics, stability, ageing and lifetime. The articles that were analysed in an earlier work, also referred to as “older dataset”, were removed from the total pile and the remaining articles, referred to as “new dataset”, were inserted in an *online database* for further analyses. The total number of article in the new dataset was 2286, out of which 303 contained actual lifetime data, while the rest only discussed the theory behind the stability issue. The 303 articles presented ageing curve for total of 983 devices, which are called data points. For the comparison, in the earlier article scanning study total of 2500 articles were scanned, which also revealed precisely 303 articles with actual experimental lifetime data. It is worth mentioning that the new dataset contains also articles from conference proceedings dating before 2013 that were not recorded in the older dataset.

2.2 Lifetime determination

The *online database* with the new articles (hosted at http://plasticphotovoltaics.org/) was shared among and analysed by different groups from consortia of the COST Actions project (http://www.cost.eu/COST_Actions). The analysis involved scanning each article individually, identifying whether the article contains experimental lifetime data, registering the reported data by filling the database with the reported sample structures, encapsulation, testing conditions and determining the lifetime from the reported ageing curves. The latter was realized by following the steps outlined in the Table 1 and Figure 1 below. A more detailed explanation is provided elsewhere [11].

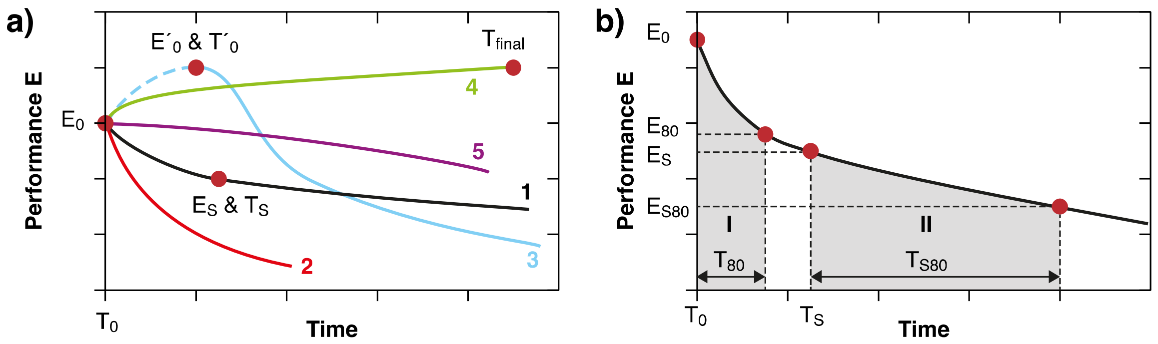


Figure 1. (a) Examples of various typical shapes of ageing curves taken from real data. (b) Example of identifying the best pair describing the stability of the sample. Reprinted with the permission from XXX.

Table 1. The list of steps for determining the lifetime marker. Reprinted with the permission from XXX.

Parameters	Method
***Determination of starting point E_0 &T_0** E_0 – initial performance T_0 – initial time	T_0 & E_0 pair is either chosen at the first measurement point or if the curve has an initial increase followed by a reduction (such as the curve 3 in Figure 1 (a)) then T_0 & E_0 is set at the maximum point.
Determination of stabilized section E_S & T_S E_S – performance at the start of stabilized section T_S – starting time of stabilized section	If after a certain point the ageing curve enters into a more stable phase (commonly observed during solar cell ageing), then a second pair of starting values T_S & E_S is identified, typically chosen at a point from where the ageing rate almost doesn't change anymore, as shown on curve 1 in Figure 1 (a).
Determination of T_{80} and T_{S80} T_{80} – time when performance reaches 80% of E_0 T_{S80} – time when performance reaches 80% of E_S	T_{80} (or if applicable T_{S80}) is determined by subtracting T_0 (or T_S) from the time when 80% of E_0 (or E_S) is reached. Figure 1 (b) highlights the areas determined by T_{80} and T_{S80}
Lifetime marker [E_0;T_{80}] or [E_S;T_{S80}]	The largest area among I and II in Figure 1 (b) (part of the curve where the sample produces the largest amount of energy) will then determine the pair that will describe the lifetime. The simple geometrical calculations reveal that the ratio of the areas of the trapezoids I and II are proportional to the ratio of the areas of the rectangles defined by the products of $E_0 \times T_{80}$ and $E_S \times T_{S80}$. Thus the lifetime marker can be mathematically identified according to these rules: if $\frac{[E_0*T_{80}]}{[E_S*T_{S8}\ ]} \geq 1$ then the marker is [E_0;T_{80}] if $\frac{[E_0*T_{80}]}{[E_S*T_{S80}]} < 1$ then the marker is [E_S;T_{S80}]

Exceptions	Exceptions are made in the following cases: • If E_S is less than half of E_0, in which case the sample is considered to have degraded before stabilization (see curve 2 in Figure 1 (a)), then [E_0;T_{80}] is chosen by default to represent the lifetime. • If the measurements has been stopped prior to reaching the 80% threshold then “$T_{final} - T_0$” or “$T_{final} - T_S$”, where T_{final} is the point of last measurement (see curve 4 in Figure 1 (a)) is chosen instead to represent the minimum possible lifetime.

The data is made publicly available at http://plasticphotovoltaics.org/lifetime-predictor.html, where an online interface can be found that allows analysing and reproducing the collected data with application of specific filters. An instruction video is additionally uploaded for navigating though the tool and the database.

3 Results and discussion

The data analysed collected from the new dataset was compared with the older dataset. The comparison revealed no significant difference in the data distribution between the two, but rather one complemented the other. The two datasets were therefore combined, which enabled better intercomparison and baselining of the lifetime distributions under different test conditions.

Figure 2 shows the increase in the device stability and quantity of the reported lifetime data in the recent years based on the combination of the two datasets. There is an obvious increase in the values in the recent years with the total number of data points reaching beyond 300, which corresponds to more than 100 articles per year (given that one article contains about 3 data points). This is a clear indication of how important the issue of lifetime has become in the recent years.

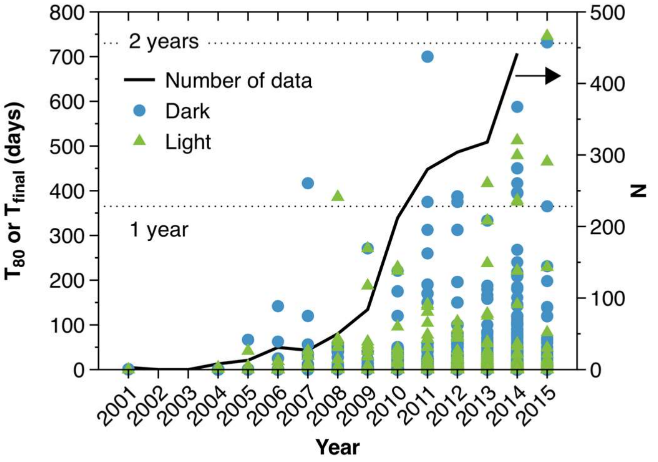


Figure 2. The scatter plot shows the T_{80} values versus the reporting year for the samples tested under light (green triangles) and in dark (blue circles). The black line shows the number of reported data-points per year until 2014.

3.1 Baseline for lifetime

The combination of the two datasets significantly increased the total amount of data points and therefore improved the statistical significance of the lifetime distributions for the samples tested under different test conditions. This enabled the possibility for establishing baselines based on such distributions. In order to do so, the data were categorized according to four groups similar to the earlier work: *group 1* and *group 2* represented the *unencapsulated* samples tested under light and in dark respectively and *group 3* and *group 4* hosted the *encapsulated* samples tested correspondingly under light and in dark. The tests under light were further distinguished by:

- indoor soaking under light source with spectrum close to AM1.5 and intensity close to 1 sun
- indoor exposure to low UV or low intensity light
- outdoor testing under real sun

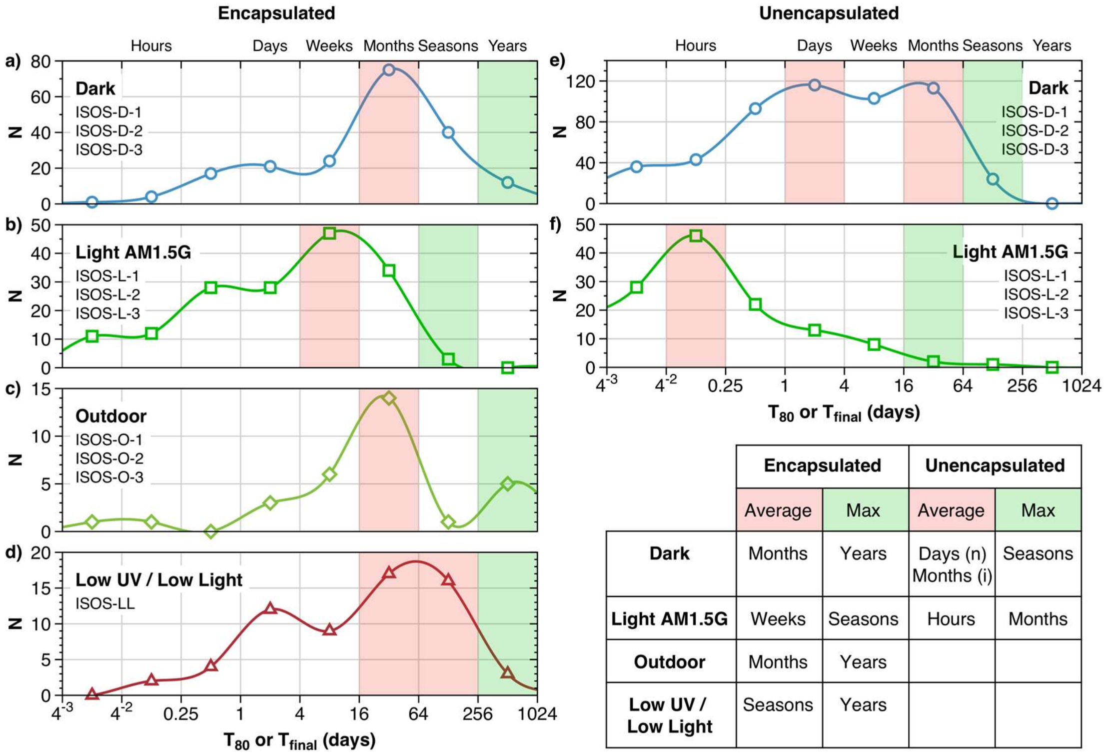
Figure 3 shows the lifetime data distribution for each test condition for the stability of the devices with and without encapsulation. Figure 3 (e), (f) and (a) represent the data from group 2, 1 and 4 respectively and Figure (b) – (d) group 3. The data is presented versus the logarithmic scale with base four similar to o-diagram reported earlier [9]. The scale is associated with the common time units shown on the top of the plots, which enables the more intuitive interpretation of the data. Each test category is also associated with the ISOS testing procedures shown in the legends. For each data distribution the average and the maximum lifetime region are defined, highlighted respectively with red and green markers. The average represents the most common lifetime values reported for OPVs, while the maximum values show the most outstanding lifetime reports. The corresponding time ranges for average and maximum are listed in the table on the right lower corner of the figure. The group 2 of unencapsulated samples tested in dark contains two average values representing normal and inverted device structures, which are discussed in the next section.

The established baselines can serve as reference points for the performance of any newly produced sample tested under given test conditions:

- If the sample outperforms the average then the sample has an improved stability
- If the performance is in the maximum region or beyond then the sample has an outstanding or record lifetime respectively

As a word of precaution, an attempt to predict the lifetime of the sample in outdoor test conditions based on the ratios of the indoor light soaking and outdoor tests may lead to erroneous results, since one is not the acceleration of the other. For simulation of the outdoor tests a more complicated set of accelerated tests will be required, such as combination of a number of ISOS test procedures. Unfortunately, the database presented in this work does not contain sufficient data for each individual ISOS test procedure at this stage, but with the gradual increase of the database the intercomparison of the data for ISOS will become possible enabling the development of the prediction tool.

Thus, the presented baselines should mainly be regarded as generic reference points for lifetime of organic photovoltaics for given test conditions according to the aforementioned grouping.



	Encapsulated		Unencapsulated	
	Average	Max	Average	Max
Dark	Months	Years	Days (n) Months (i)	Seasons
Light AM1.5G	Weeks	Seasons	Hours	Months
Outdoor	Months	Years		
Low UV / Low Light	Seasons	Years		

Figure 3. Baselines of the lifetime of OPVs tested under different ageing conditions for encapsulated (left plots) and unencapsulated (right plots) samples. The plots represent the number of data points against the time in days represented in logarithmic scale with base 4. The scale is associated with the common time units shown above the plots. The average and maximum lifetime values are highlighted in red and green and are listed in the table on the right lower corner. For unencapsulated samples tested in the dark there are two distinct peaks and thus to average values of days and months representing normal and inverted structures (see section 3.2). The test conditions are associated with but not limited to the ISOS test conditions.

3.2 Normal vs inverted structures

In Figure 3 (e) the unencapsulated samples tested in the dark show two distinct peaks. These correspond to device with normal (also known as conventional) and inverted architectures. The former typically employs aluminium back electrode, while the latter has Ag or Au based electrode. Figure 4 shows the comparison of the conventional and inverted devices for samples with and without encapsulation. From the figure it is apparent that there is a significant difference in the intrinsic stability of the normal and inverted structures, which is less pronounced in the case of the encapsulated samples. It has been established earlier that the normal structures are significantly less resistant towards the moisture due to the high sensitivity of the aluminium [12–14] and therefore show inferior stability when tested in the dark. In the case of encapsulation the sample becomes protected from the humid environment and therefore the reaction of the electrode with moisture is significantly reduced. In the indoor light tests, there is no obvious difference in the stability of the two structures, since the heat produced by the light source creates rather dry environment around the sample diminishing the effect of humidity. As a result the encapsulation of the normal structure devices has a major impact on the stability, while in the case of inverted structures the role of encapsulation does not seem to be significant when the samples are stored in dark as can be seen in Figure 4.

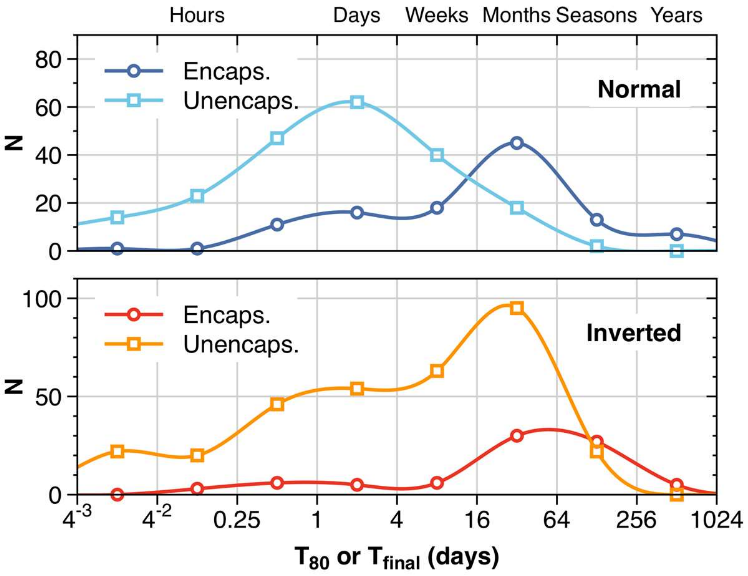


Figure 4. The lifetime distribution of the sample with normal (top) and inverted (bottom) structures tested in the dark. The dark and light curves correspond to encapsulated and unencapsulated samples respectively.

3.3 Winning Structures

In the older dataset collected from the earlier article scanning project there was a number of device architectures outlined with reported best intrinsic stabilities (unencapsulated samples). Similarly, in the new dataset a number of reports with samples of outstanding intrinsic stability were registered, which are outlined in the Table 2 below. The table highlights the structures of the reported samples tested under light or in dark and their corresponding lifetime and efficiency values. The most impressive report is the sample tested under light that has showed a lifetime of 96 days [15]. Unfortunately, the details of the top electrode configuration were not reported, but it was stated that it contained a combination of different metals. It is worth mentioning also that one of the samples tested in the dark that showed an outstanding stability of 120 days, was produced in a roll-to-roll compatible process utilizing coating and printing techniques [9]. Nevertheless, despite a number of reports of impressive intrinsic stability, producing samples in a roll-to-roll compatible process with sufficient stability under light test presents a serious challenge that still needs to be addressed [16].

Table 2. The structure and performance parameters of unencapsulated devices tested in dark and under light. The active layer of all the materials is identical and consists of P3HT:PCBM[60].

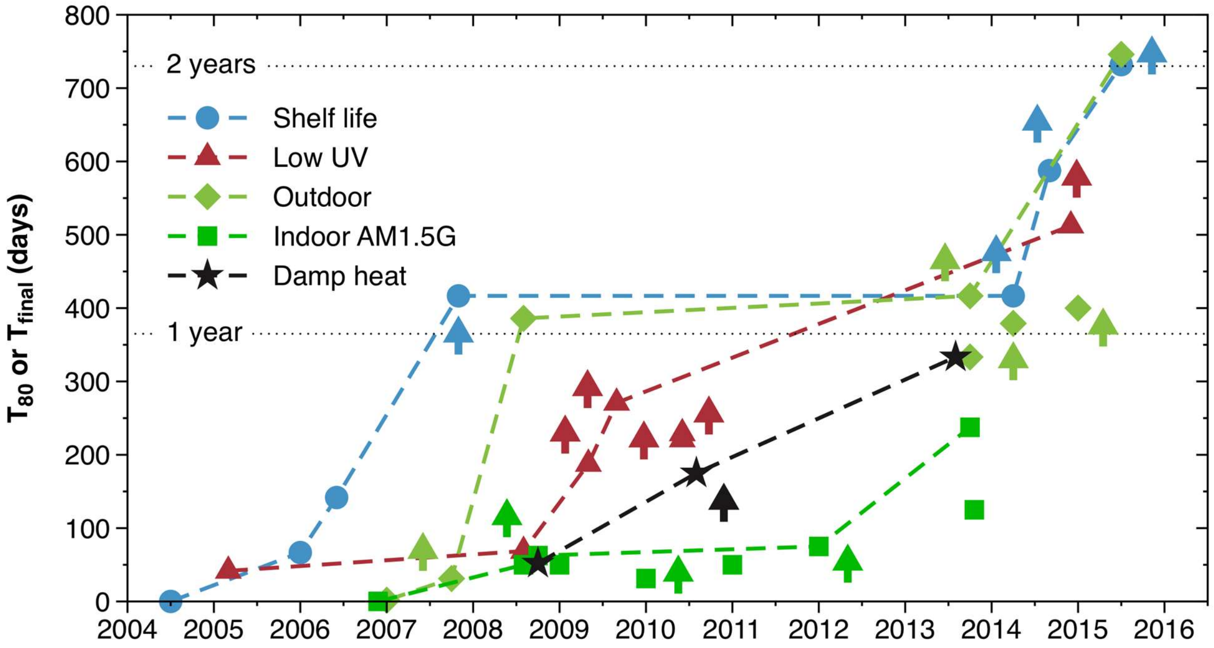
	Dark			**Light**	
Back Electrode	**Ag / Ag+Al / Ag**	**Ag grid**	**Al**	**Multilayer metal electrode**	**Ag**
Transport Layer 2	***MoOx / PEDOT:PSS / None**	**PEDOT:PSS**	**Cs2CO3**	**PEDOT:PSS**	**MoOx**
Active Layer	**P3HT:PCBM**	**P3HT:PCBM**	**P3HT:PCBM**	**P3HT:PCBM**	**P3HT:PCBM**
Transport Layer 1	**TiOx / ZnOx / ZnOx**	**PEDOT:PSS + ZnOx**	****Other**	**ZnOx**	**ZnOx**
Front Electrode	**ITO**	**Ag grid**	**ITO**	**ITO**	**ITO**
Substrate	**Glass**	**PET**	**Glass**	**Glass**	**Glass**
Structure	**Inverted**	**Inverted**	**Normal**	**Inverted**	**Inverted**
PCE (%)	**3.7 / 3.5 / 2.5**	**0.93**	**3.6**	**1.9**	**2.85**
Lifetime (days)	**198 / 187 / 146**	**120**	**100**	**96**	**17.5**
Reference	[17],[18],[19]	[9]	[20]	[15]	[21]

* MoOx modified with Nafion

** Phenothiazine, 4-phenothiazin-10-yl-anisole (APS)

3.4 Plot of the record lifetimes

From the previous report a so called lifetime progress diagram was presented, which highlighted the best reported lifetimes of organic solar cells tested under different test conditions. The diagram has been updated by additions from the new dataset and is presented in Figure 5. The references of the reports are provided in the table below the image.



Indoor AM1.5G			Shelf Life			Outdoor			Low UV			Damp Heat		
PCE (%)	Lifetime (days)	Ref.	PCE (%)	Lifetime (days)	Ref.	PCE (%)	Lifetime (days)	Ref.	PCE (%)	Lifetime (days)	Ref.	PCE (%)	Lifetime (days)	Ref.
1.08	0.083	[22]	0.8	0.042	[23]	0.0024	2	[24]	2.5	42	[25]	NA	52	[26]
NA	50	[27]	0.035	67	[28]	4.2	31	[29]	4.1	69	[30]	4.4	175*	[31]
NA	63	[26]	0.16	142	[32]	NA	386	[27]	2.32	188	[33]	3	333	[34]
1.09	50	[35]	2.8	417	[36]	1.43	417	[37]	5.9	271	[38]			
3.54	31	[39]	1.27	417	[40]	1.43	333	[37]	2.7	229	[41]			
NA	50	[42]	6.05	587.5	[43]	NA	379	[40]	6.07	221	[44]			
2.1	75	[45]	1.06	732	[46]	1.42	400	[47]	2.7	513	[48]			
3.42	125	[49]				1.11	746	[46]						
2.59	238	[50]												

**Not compatible with ISOS-D-3 conditions: Tested at 25 °C air temperature*

Figure 5. The best reported lifetime for each year

4 Conclusions and future perspective

This article presented the results of the article analysis published in literature related to the stability of organic solar cells reported in the recent years. The progress in the number of reports per year dealing with the lifetime of OPVs was shown, which asserted the ever increasing interest towards resolving the stability issue of this technology. From the large dataset baselines were determined for the lifetime of OPVs, tested under different conditions, which can serve as a reference point for determining whether a newly reported data has an improved or record lifetime compared to commonly reported values. In addition, a list of devices with outstanding intrinsic stability was highlighted together with the detailed analysis of their structures. The updated version of the diagram of the record stabilities was presented as well. The work constitutes a step forward towards ongoing process of the development of a prediction tool for reliably determining the sample durability. The major challenge is the significant lack of experimental data for each individual ISOS testing condition and in particular for the outdoor tests, which hampers the development of the tool and therefore the work will continue towards generating and collecting more outdoor data.

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