



An effective method of predicting perovskite solar cell lifetime—Case study on planar $\text{CH}_3\text{NH}_3\text{PbI}_3$ and $\text{HC}(\text{NH}_2)_2\text{PbI}_3$ perovskite solar cells and hole transfer materials of spiro-OMeTAD and PTAA

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ABSTRACT

As stability of perovskite solar cells remains a significant research topic, it is important to be able to predict the long-term stability of any new kinds of perovskite solar cells when new perovskite absorber materials or transport layers or new cell structures are being demonstrated. This work reports a reliable method of determining degradation rate which is resulted from thermal stress. By incorporating three kinds of accelerated tests, the activation energy for photo-thermally driven degradation processes of perovskites solar cells was determined, which is then used to predict its long-term stability using an Arrhenius equation. In addition, thermal stability of $\text{CH}_3\text{NH}_3\text{PbI}_3$, $\text{HC}(\text{NH}_2)_2\text{PbI}_3$, PTAA (poly[bis(4-phenyl)(2,4,6-trimethyl phenyl)amine]) and Spiro-OMeTAD (2,2',7,7'-Tetrakis[N,N-di(4-methoxyphenyl)amino]-9,9'-spirobifluorene) are studied. The thermal stability of a planar $\text{HC}(\text{NH}_2)_2\text{PbI}_3$ /PTAA device is better than a planar $\text{HC}(\text{NH}_2)_2\text{PbI}_3$ /Spiro-OMeTAD device which in turn is better than a planar $\text{CH}_3\text{NH}_3\text{PbI}_3$ /Spiro-OMeTAD device due to better thermal stability of $\text{HC}(\text{NH}_2)_2\text{PbI}_3$ and PTAA. It is predicted that a planar $\text{HC}(\text{NH}_2)_2\text{PbI}_3$ /PTAA device can have a lifetime of more than 3 years (or 1.5 years) at room temperature if 50% (or 25%) drop in power output can be tolerated. While these lifetimes are specific to perovskite material chosen, preparation method and solar cell design, the lifetime prediction method reported here can be verified experimentally. Therefore, the lifetime calculation method developed in this work is a quick and useful tool for determining the relative stability of a perovskite device especially when comparing the merits of different cell structure designs.

1. Introduction

Recently, organic–inorganic halide perovskite solar cells have emerged as a promising alternative to existing photovoltaic technologies due to its ease of fabrication and excellent absorption and charge transport properties [1]. However, the most important challenge perovskite solar cell faces its instability given that the incumbent crystalline silicon (c-Si) solar cell technology is abundant and proven with module guaranteed-lifetime of more than 20 years [2,3]. Instability of perovskite solar cell is well known upon exposure to air with moisture and is most severe at elevated temperatures. Instability from organic charge transport layers [4], highly corrosive perovskite under moisture [5–8], metal contacts [8,9] have been reported. Methods to protect the perovskite solar cell from moisture have been proposed showing encouraging results under high humidity [10–13].

Indium tin oxide (ITO) [14] and carbon electrodes [15] as alternatives to metal electrodes have been shown to provide better thermal stability. In terms of perovskite absorber layers, it has been reported that cesium based perovskites have better thermal stability [16–18] than $\text{HC}(\text{NH}_2)_2$ (FA) which in turn is better [19,20] than CH_3NH_3 (MA) perovskites [21,22]. Most recently, two-dimensional perovskite devices with modest conversion efficiencies have also been shown to have excellent stability in [23]. Inorganics metal-oxide carrier blocking and transport layers have also been demonstrated to provide good stability [24] compared to devices using organic transport layers [24,25]. Given the potential solutions available with many yet to be explored and the limited lifetimes (compared to the incumbent c-Si solar cell technology), of even the most stable devices reported, there remains opportunities for further stability study of perovskite solar cells.

Here we report a quick method which can be applied to any

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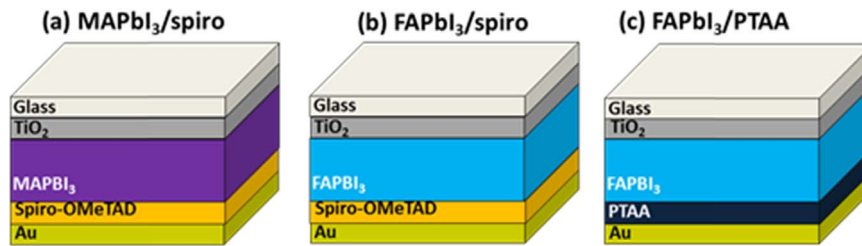


Fig. 1. Structures of perovskite solar cells studied in this work.

perovskite solar cell structure to determine its degradation rate and to calculate its lifetime from the degradation activation energy. We apply this method to three types of planar perovskite solar cells (Fig. 1): (1) FTO Glass/c-TiO₂/MAPbI₃/spiroOMeTAD/Au, (2) FTO Glass/c-TiO₂/FAPbI₃/spiroOMeTAD/Au, and (3) FTO Glass/c-TiO₂/FAPbI₃/PTAA/Au to determine the activation energy for degradation. Planar instead of meso-porous structure is chosen in this work due to their simplicity [26,27] which would explicate the intrinsic perovskite materials stability and therefore the viability of planar structure. The glass/c-TiO₂/perovskite/HTM/Au structure is chosen because it is one of the more popular planar structures studied that can be used as a baseline or control structure although there is scope for alternative ETMs such as tin oxide (SnO₂) [28,29].

2. Material and methods

2.1. Experiments

The details of material synthesis, test structure and solar device fabrications [30,31] and encapsulations are given in [Supplementary Information](#). Fig. 1 shows the three solar device structures for this study. The average power conversion efficiencies (PCE) of the devices are 13%, (see Fig. S1 in [Supplementary Information](#)).

The accelerated thermal tests of the perovskite test structures and perovskite solar cells were conducted by heating continuously on a hot plate at each temperature in a glovebox (H₂O and O₂ levels < 1 ppm) monitored by K-type thermocouple. We conducted acceleration tests on the perovskite solar cells by dividing them into three groups. These groups were exposed to three different thermal stresses. These include: (a) 45 °C, (b) 65 °C and (c) 85 °C. A hot plate was used to apply thermal stress and the temperature of samples was monitored by K-type thermocouple. The electrical performance of each sample was measured every 24 h. We determined the time to measured or extrapolated 20% reduction in the initial maximum power output at all samples. Degradation rate (R_D , %/h) is the inverse of the time to 20% reduction at a given condition.

The effects of thermal stress on the morphology and structure of the perovskite and hole transport materials (HTM) are analysed by characterising the test structure through X-ray diffraction (XRD) measurement (to investigate the crystallinity), scanning electron microscopy (SEM) (to investigate the film quality), and optical microscopy such as the use of cross polarized imaging which allows the imaging of amorphous phase (dark) and crystals (bright) in the HTM. Details of the tools and methods are given in [Supporting Information](#). For the encapsulated PV devices, electrical performance was measured every 24 h.

2.2. Methods for predicting long-term stability

The degradation rate (%/h) is the inverse of the mean time to failure at a given condition. In order to determine the degradation rate (R_D) using an Arrhenius model [32], we measured the time taken for the initial maximum power of all samples to diminish by 20%. The decreasing rate of the maximum power is related to the temperature in this model, as follows:

$$R_D = A \exp\left(\frac{-E_a}{kT}\right) \quad (1)$$

where, E_a is the thermal activation energy of the degradation process (eV), k is the Boltzmann constant (8.62×10^{-5} eV/K), T is the temperature (°K), and A is a constant which can be found from the intercept ($\ln(A)$) in a $\ln(R_D)$ vs $1/kT$ plot and the slope will be $-E_a$.

It is established that the degradation (D) of the perovskite solar cells due to thermal stress over a period of time t can be computed by Eq. (2) [32]

$$D = R_D \times t = A \exp\left(\frac{-E_a}{kT}\right) \times t \quad (2)$$

If 20% of PCE drop can be tolerated, the time to 20% drop in PCE of each perovskite solar cell can be determined at different temperatures from Eq. (3)

$$t = \frac{20}{A \exp\left(\frac{-E_a}{kT}\right)} \quad (3)$$

In order to verify the degradation model of Eq. (2), degradations of the 3 types of encapsulated perovskite solar cells at 25 °C over 500 h in a glove box were measured (PCE's measured every 120 h) and modelled.

3. Results and discussion

The PCE degradations and R_D 's of the three types of perovskite solar cells are plotted in Fig. 2. In both our experience and in published work [33–35], the degradation curve flattens over time after initial degradations. This means a linear degradation rate is a more conservative estimate that likely underestimates the predicted lifetimes than in the case if we also consider the exponential degradation rate [36]. The V_{OC} , J_{SC} and FF of these devices measured during thermal stress tests are also shown in Fig. S2 to S4 in [supplementary information](#). It is evident that MAPbI₃/spiroOMeTAD cell is the least stable amongst the three types of perovskite solar cell tested. In particular, it degrades most rapidly at 85 °C and loses 20% of power output after only 48 h at temperatures above 65 °C. Stability of a perovskite solar cell improves (1) when MAPbI₃ is replaced with FAPbI₃ and (2) spiroOMeTAD is replaced with PTAA.

To understand the degradation mechanisms associated with the different types of perovskite absorbers and HTM's, characterisations of the test structures are carried out that reveal the effects of thermal stress as discussed below.

Fig. 3(a) and (b) show the XRD patterns of MAPbI₃ or FAPbI₃, respectively, before and after thermal stress at 85 °C. The MAPbI₃ has two peaks at 14.1° and 28.36°, indexed to the (110) and (220) planes, respectively. The (110) peak gradually disappears with thermal stress while the peak of PbI₂, (100), at 12.64° appears after 48 h at 85 °C. This is in agreement with the reported result [21]. Note the degradation of MAPbI₃ only happens at 85 °C as negligible changes in the XRD or in the SEM can be detected for MAPbI₃ heated at 45 °C or 65 °C, see Fig. S5 and S6. For the FAPbI₃ film, there is no pronounced emergence of the PbI₂ peak. The SEM images of the FAPbI₃ film also shows

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