

Machine Learning Models Forecasting Optoelectronic Service Lifetimes of Low-
Bandgap Perovskite Solar Cells

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Abstract

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Accurately predicting the lifetime of perovskite solar cells (PSCs) is essential for their successful commercialization. Mixed Sn-Pb halide perovskites are promising materials because of their relatively low bandgap (~ 1.2 - 1.3 eV) making them a strong candidate for use as the absorber layer in single-junction solar cells or as the low-bandgap subcell absorber in all perovskite tandem solar cells. However, this composition suffers from stability issues when exposed to environmental conditions. By collecting experimental data from accelerated degradation, we learn that efficiency losses are driven by short-circuit current and indicate that the loss in perovskite material is a major contributor to degradation. Additionally, we have identified significant correlations between device lifetime and the perovskite degradation rate (derived from previous studies), partial pressure of water, and early time derivative of specific device measurements. To predict the lifetime of PSCs, machine learning models were trained using a menu of features that were split into 3 distinct categories: (1) *a priori* known features, (2) initial device measurements, and (3) measurements of

the initial rates of change of device parameters. With a dataset consisting of 39 degradation experiments, trained models reveal that the derived degradation rate of the $\text{FA}_{0.75}\text{Cs}_{0.25}\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_3$ perovskite material is a valuable feature for forecasting PSC lifetime and was selected as the most dominant feature in the best performing models which demonstrate a median test error of 26%. These findings underscore the importance of quantitative measurements of perovskite degradation in accurately forecasting the lifetime of PSCs and highlight the utility of integrating such metrics into predictive models.

1. Introduction

In the face of worsening climate changes due to global warming, the demand for cost-effective renewable energy source has escalated rapidly. Due to its ability for clean energy generation, photovoltaic (PV) technology has gained significant attention. A critical metric for evaluating the feasibility of PV systems commercialization is the levelized cost of energy (LCOE). LCOE depends on net total cost of producing and operating the power generating asset and the total electricity generation over its lifetime. The PV market is currently monopolized by crystalline silicon solar cells reaching a peak efficiency of 22.2% in commercial products¹ and a record efficiency of 26.6% in research cells.² However, LCOE for crystalline silicon solar cells averages around \$0.11 per kWh when compared to natural gas which is \$0.05 per kWh.³ In order for PV technology to be competitive, US Department of Energy (DOE) Solar Energy Technology Office (SETO) has launched the SunShot 2030 initiative which aims for an LCOE of \$0.03/kWh, which is considered achievable with solar panels that have a total area efficiency of 25%, a module cost of \$0.10/Wdc, and a lifespan exceeding 30 years. Nonetheless, the high manufacturing cost and the limited theoretical maximum PCE of 29.4% for silicon solar cells⁴ present significant challenges to reaching these goals.

Perovskite solar cells (PSCs) have been in discussion as a potential replacement for silicon based solar cell for a while due to their low production cost,⁵ comparable efficiency (record power conversion efficiency of 26.1%),⁶ flexible processing compatibility,^{7,8} and highly tunable bandgaps.⁹ However, the stability of PSCs continue to be a main concern which limits their potential for commercialization.^{10,11} Enhancements in PSC stability are often achieved through fabrication method optimization¹², perovskite compositional tuning^{13,14}, or the use of

encapsulation techniques.^{15,16} While there are many standard testing conditions for solar cell and module stability, such as the IEC-61215 damp heat test issued by the International Electrotechnical Commission (IEC), which are used to test Si-based and GaAs-based photovoltaics (PV), studies have shown that many of these tests are unsuitable for PSCs which exhibits unique behaviors such as hysteresis and degradation due to the effects of reverse bias.¹⁷ While many studies have tested the stability of PSCs, there is a lack of unified stability and accelerated aging test among different groups that would allow for an unbiased assessment of stability.¹⁸ Our group proposed to develop forecasting models of PSC performance through machine learning techniques that incorporate our knowledge of the unique issues that plague the stability of perovskite semiconductor based devices, particularly the degradation kinetics of perovskite materials.

Our group has performed extensive studies aiming to elucidate the specific decomposition mechanisms of methylammonium lead triiodide perovskite (MAPbI₃) thin films. The kinetics governing the early-time reaction rate of MAPbI₃ loss were discovered using optical absorbance. By assessing this early time degradation across a wide range of conditions of different light intensity, temperature, oxygen concentration, and relative humidity (*RH*), we were able to derive a rate expression that could accurately describe the degradation rate quite accurately with a $R^2 = 0.75$ (predicted from rate equation versus observed from experimental measurements) and median relative error of 10%.¹⁹ Studies on MAPbI₃ devices were then carried out where a machine learning model was developed to help predict the time required for the power conversion efficiency to drop to 80% of its initial value (*T*80). The incorporation of the theory-based physiochemical feature derived from a model of the chemical reaction kinetics of unencapsulated MAPbI₃ films was particularly valuable

for prediction, suggesting that perovskite absorber material loss is a crucially important aspect of PSC *PCE* loss over time. This physiochemical feature was selected as the first or second most dominant feature within the best performing models.²⁰ Based on the study of MAPbI₃ perovskite, we believe that a similar methodology can be used to determine the kinetically limited rate expression of bare perovskite film, elucidate the reaction mechanisms of device degradation, as well as develop models for different perovskite compositions to predict the *T80* of PSCs.

In this proposed study, my focus will be on mixed Sn-Pb halide perovskites because of their relatively low bandgap (~1.2-1.3 eV) which makes them a strong candidate for use as the absorber layer in single-junction solar cells or as the low-bandgap subcell absorber in all perovskite tandem solar cells. Despite their strong potential, Sn-based perovskite suffers from extreme instability due to the quick oxidation of Sn²⁺ into Sn⁴⁺ making it important to understand its degradation mechanism. The composition of FA_{0.75}Cs_{0.25}Pb_{0.5}Sn_{0.5}I₃ was selected for previous studies on bare films. Similar to the studies conducted on MAPbI₃ films, in-situ measurements of optical absorbance (calculated from transmittance (*T*)) were used to derive the kinetic rate expressions of the perovskite degradation where two major degradation pathways were discovered, dry oxidation and water-accelerated oxidation. A simple machine learning model was then developed to predict the time for diffusion length to decrease to 80% of its initial value (*t_{L_D,80}*), this is because *PCE* is highly correlated with the decay of short-circuit current density (*J_{SC}*), and *J_{SC}* is proportional to diffusion length. The model performed well with a median test error of ~32.6%, where the chemical decomposition rate expression was chosen as the most dominant feature, again highlighting the utility of the kinetic rate expression in performance-based forecasting models of perovskite materials.²¹

Following the research on bare films, the goal of this study is to figure out the degradation mechanism of $\text{FA}_{0.75}\text{Cs}_{0.25}\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_3$ perovskite devices as well as develop a machine learning model which would be able to forecast the T_{80} of the devices following a similar methodology as the studies done on MAPbI_3 devices.

2. Experimental

2.1 Device Fabrication

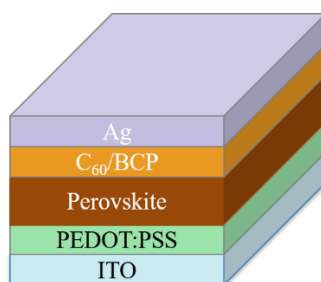


Figure 1 Device architecture, note that layer thicknesses are not to scale.

The perovskite composition chosen is a mixed Sn-Pb perovskite with the composition of $\text{FA}_{0.75}\text{Cs}_{0.25}\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_3$. Devices are fabricated in the configuration of ITO/PEDOT:PSS/ $\text{FA}_{0.75}\text{Cs}_{0.25}\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_3$ /C₆₀/BCP/Ag as shown in Figure 1. In a nitrogen filled glovebox, precursor inks are prepared by dissolving stoichiometric amounts of tin(II) iodide (SnI_2 , Sigma Aldrich, perovskite-grade), formamidinium iodide (FAI, Greatcell Solar Materials, >99.99%), lead(II) iodide (PbI_2 , TCI AMERICA, 99.99%, trace metals basis), cesium iodide (CsI, Fisher Scientific, 99.998%), and tin(II) fluoride (SnF_2 , Sigma Aldrich, 99%) at 1.5 M concentration in a 4:1 vol:vol mixture of N,N-dimethylformamide (DMF, Sigma Aldrich anhydrous grade) and dimethyl sulfoxide (DMSO, Sigma Aldrich anhydrous grade). The perovskite solution is then left to dissolve at 30 °C for at least 2 hrs. Pre-patterned indium tin oxide (ITO) glass substrates (1.5×1.5 cm, $15 \, \Omega \, \text{sq}^{-1}$, Yingkou Shangneng

Photoelectric Material Co.) are cleaned by sonication in Alconox detergent solution, deionized water, acetone, and isopropanol for 10 min in each solution, then blow-dried with clean N₂ gas and subsequently treated with an O₂ plasma for 10 min. After cleaning, 50 µL of diluted 1:3 vol:vol mixture of poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS, Heraeus) and methanol (CH₃OH, Fisher Scientific, 99.8%) is spin coated at 4000 rpm for 20 s, followed by annealing on a ceramic hot plate at 140 °C for 10 min. The films are immediately transferred into a nitrogen-filled glovebox for perovskite deposition. 5 µg of tin powder is added to the perovskite ink and stirred for 10 min enabling the reduction of Sn⁴⁺ to Sn²⁺ before deposition. The solution is then filtered with a 0.2 µm PTFE filter and 50 µL is spin coated on each substrate at 5000 rpm for 40 s. In the last 12 seconds during spin coating, a 120 µL anisole anit-solvent wash is dropped onto the film over 2 seconds. The films are then annealed on a ceramic hot plate at 100 °C for 10 min. During perovskite deposition, the glovebox was continuously purged with nitrogen to avoid solvent buildup. Samples are swiftly transferred to a thermal evaporator (Angstrom Engineering Inc.) where the following layers and its corresponding thickness are deposited through thermal evaporation deposition: 40 nm of fullerene (C₆₀, Sigma Aldrich, 99.5%), 7 nm of bathocuproine (BCP, Sigma Aldrich, sublimed grade), and 100 nm of silver (Ag, Kurt Lesker, 99.99%). The active working area of the fabricated device is 0.06 cm².

2.2 Device Characterization

J-V curves of all devices are measured under simulated AM1.5 1 Sun illumination with 0.1 V/s sweep rate for forward and reverse sweeps with a mask area of ~0.0453 cm². The light source is an Oriel VeraSol-2 AAA Solar Simulator. An OSI Optoelectronics UV-100DQ Si photodiode was used to set the lamp intensity to equal

the integrated above-bandgap AM1.5GT photon flux for a 1.27 eV bandgap material. Measurements for J_{SC} , V_{OC} , FF , V_{MPP} (maximum power point voltage), and power conversion efficiency (PCE) is calculated by the following equation:

$$PCE = \frac{V_{OC} \times J_{SC} \times FF}{P_{in}} \quad (1)$$

Where V_{OC} is the open circuit voltage (V), J_{SC} is the short-circuit current density (mA/cm²), FF is the fill factor (unitless), P_{in} is the input power (mW/cm²).

2.3 Accelerated Degradation

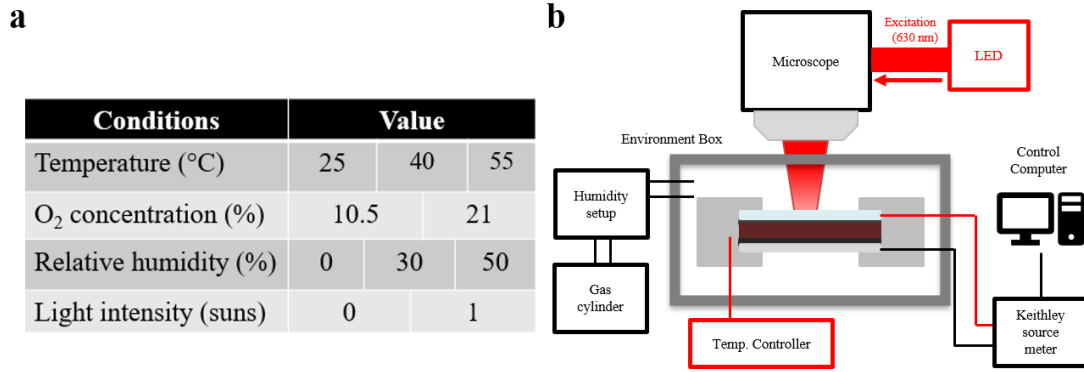


Figure 2 (a) Inserted table list the conditions used for degradation experiment. (b) Experimental setup for accelerated degradation.

For degradation experiments, devices prepared as described above are placed under conditions with varying temperature, O₂ concentration, relative humidity, and light intensity listed in the table in Figure 1a. To control the environmental conditions, devices are placed in a homemade environmental test chamber, outfitted with an equipped with cartridge heaters and closed-loop thermocouple temperature control, gas ports, electrical probes, and a window above the sample to allow for illumination as illustrated in Figure 2b. The atmosphere was controlled by connecting a cylinder of the desired gas (N₂ or dry air and through a H₂O/glycerol bath if humid gas is needed)

to the gas port on the environment chamber. Relative humidity is adjusted by changing the ratio of H₂O to glycerol in the bath and verified by a Fisher Scientific Traceable™ hygrometer. The desired gas is flowed into the chamber at 2.0 L/min using twin Omega FMA5400/5500 mass flow controllers connected to the gas cylinders. A calibrated 630 nm LED is used as the light source in a metallurgical upright microscope (Olympus BX53M, equipped with a 5X Mitutoyo Plan Apo NIR HR objective lens), and the intensity is adjusted based on 1.27 eV band gap to obtain the desired 1 sun equivalent flux of above-band gap photons under the AM1.5G spectrum. The microscope was set up to illuminate the sample and detect re-emitted radiation in reflection geometry, allowing photoluminescence (PL) and dark field (DF) images to be captured by placing an appropriate filter cube in the path. During degradation experiment, the device's electrical measurement such as V_{OC} , I_{SC} , voltage at max power point (V_{MPP}), and current at max power point (I_{MPP}) are recorded using a Keithley 2400 source/measure unit and recorded every 8 minutes using a custom-written Python code. The FF and PCE can then be calculated from collected measurement.

2.4 Feature Selection Methods and Machine Learning Modeling

Feature calculation is done with Python code and the results are processed through several different feature selection methods which are as followed: lasso (ℓ_1)²², adaptive ℓ_1 (ℓ_1^{AD})²³, iteratively reweighted ℓ_1 (ℓ_1^{IR})²⁴, best-subset (ℓ_0)²⁵, a hybrid of ℓ_0 and ridge regression ($\ell_0\ell_2$)²⁶, Fast Correlation Based Filter ($FCBF$)²⁷, orthogonal matching pursuit (OMP)²⁸, iterative sure-independence screening ($ISIS$)²⁹, and *Boruta*³⁰. The final machine learning model is created using the selected features and its coefficients using a simple linear model.

ℓ_1 , ℓ_1^{AD} , ℓ_1^{IR} , ℓ_0 , and $\ell_0\ell_2$ seek to minimize prediction error (mPE) by minimizing the sum of ordinary-least-squares (OLS) and a penalty term which controls the sparsity. ℓ_1 minimizes a loss function $\mathcal{L}_{\ell_1}(\beta)$ shown in Eq 2.

$$\mathcal{L}_{\ell_1}(\beta) = \sum_{i=1}^N \left[y_i - \left(\beta_0 + \sum_{j=1}^p \beta_j x_{ij} \right) \right]^2 + \lambda \cdot \sum_{j=1}^p |\beta_j| \quad (2)$$

The first term is the OLS and the latter is the penalty including a hyperparameter (λ) controlling the sparsity (s) and the ℓ_1 penalty term. x_{ij} is the predictor variables, y_i is the responses, p is the feature variables, and β_j is the coefficient. ℓ_1^{AD} and ℓ_1^{IR} are weighted ℓ_1 methods where the ℓ_1 penalty term becomes

$$\sum_{j=1}^p w_j |\beta_j| \quad (3)$$

For ℓ_1^{AD} , coefficient is obtained from an OLS fit (β^{ols}) where the weight is calculated using the following equation:

$$w_j = \frac{1}{|\beta_j^{ols}|^\gamma + \epsilon} \quad (4)$$

γ is a tunable hyperparameter and ϵ is a small constant to avoid division by zero. For ℓ_1^{IR} , coefficient β^* is first determined using traditional ℓ_1 , the weight w is obtained from:

$$w_j = \frac{1}{|\beta_j^*|^\gamma + \epsilon} \quad (5)$$

w is then used to fit a weighted ℓ_1 and a new w is obtained from the fitted β^* where this process is repeated a predetermined number of times. ℓ_0 solves a discrete optimization problem by choosing the best-fitting subset of features for a given

sparsity (s). It seeks to minimize the OLS term same as above with a ℓ_0 penalty constraint such that

$$\sum_{j=1}^p \begin{cases} 1 & \text{if } \beta_j \neq 0 \\ 0 & \text{if } \beta_j = 0 \end{cases} = s \quad (6)$$

$\ell_0\ell_2$ is a hybrid version of ℓ_0 and ridge regression which augment discrete subset selection with shrinkage where it minimizes a loss function $\mathcal{L}_{\ell_0\ell_2}(\beta)$ shown in Eq. 7 with a ℓ_2 penalty term along with a ℓ_0 penalty constraint as in Eq. 6.

$$\mathcal{L}_{\ell_0\ell_2}(\beta) = \sum_{i=1}^N \left[y_i - \left(\beta_0 + \sum_{j=1}^p \beta_j x_{ij} \right) \right]^2 + \lambda \cdot \sum_{j=1}^p |\beta_j|^2 \quad (7)$$

FCBF aims for minimization of redundancy and maximization of relevance (mRMR). It ranks all the features based on mutual information (MI), a measure of non-linear correlation, shared with the target variable. Lower ranked features that share high MI with higher ranked features are then removed to increase sparsity.

OMP and *ISIS* work similarly where they seek to reduce the residual, the differences between observed and predicted values of data, below a certain threshold or a maximum iteration is met. *OMP* starts by selecting the most correlated feature to predict the target variable and obtain a residual. In each iteration, it selects the feature, which are different from the ones already selected, with the highest correlation with the residual and updates its prediction obtaining a new residual. It iterates this loop until it reaches a set maximum iteration or the residual become sufficiently small. *ISIS* performs an initial screening of the feature list by employing sure independence screening (SIS), this can filter out nonrelevant features which can be done by calculating the Pearson correlation between features and the target variable. Features

with low correlation are discarded. ℓ_1 method is then performed on the features that pass the initial screening and a prediction model is created along with the residual. In each iteration, SIS is applied to obtain a new set of features with the residual being the new target. This is looped until reaching a predetermined number of iteration or the residual falls below a threshold.

Boruta method minimizes the false-discovery rate which is the rate that selected features are truly null. It achieves this by first creating a shadow feature list, this feature list is a clone of the original data but the data within are shuffled, destroying any correlation that the original data has with the target variable. A random-forest is then fit over the augmented dataset to evaluate feature importance. Hits are assigned to true features when deemed more important than all shadow features, this process is repeated M times. A binomial distribution of hits for each true feature over M repeats is obtained and features with high hit probability is chosen.

3. Results and Discussion

3.1 Device Performance

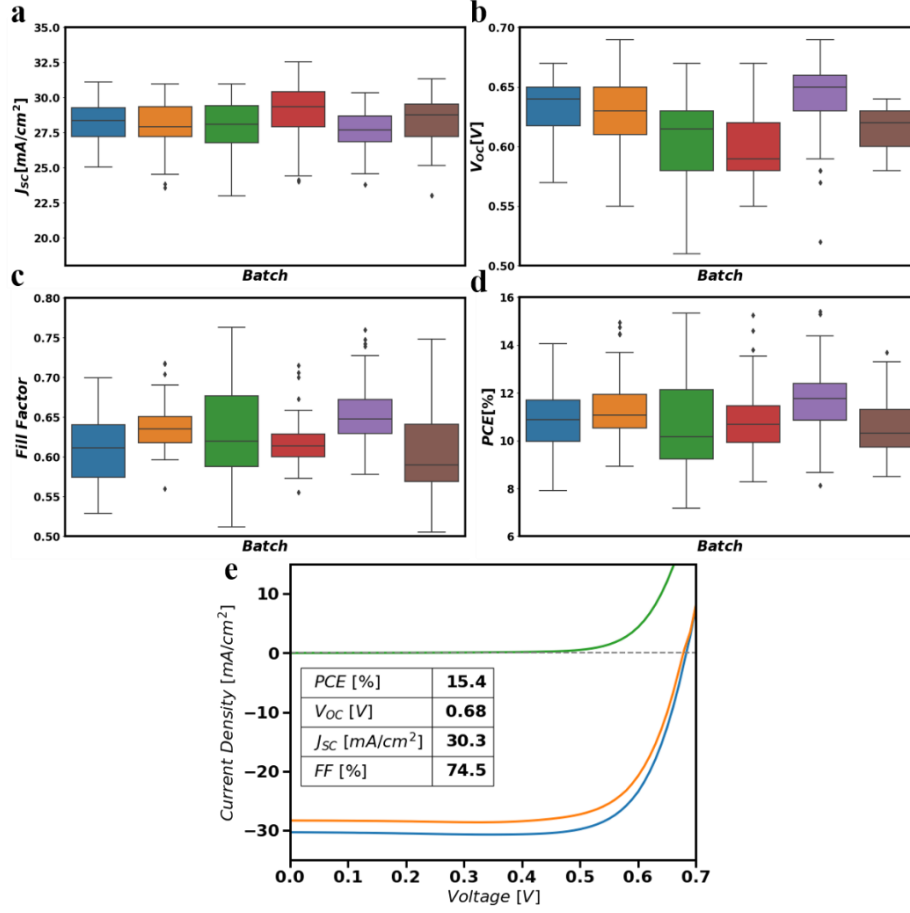


Figure 3 Device performance of the (a) short-circuit current density (b) open-circuit voltage (c) fill factor, and (d) power conversion efficiency from several batches. (e) J-V measurement of a champion device.

The typical performance of the effective devices is summarized in Figure 3a, 3b, 3c, and 3d, with the performance parameters spanning the following ranges: the short-circuit current density (J_{sc}) varies from 25 to 32 mA/cm^2 , the open-circuit voltage (V_{oc}) ranges from 0.6 to 0.65 V, the fill factor (FF) is between 0.58 and 0.67, and the PCE falls within 10 to 12%. Additionally, the current-voltage ($J - V$) characteristics of a champion device from recent batches are depicted in Figure 3e.

3.2 Accelerated Degradation of Devices

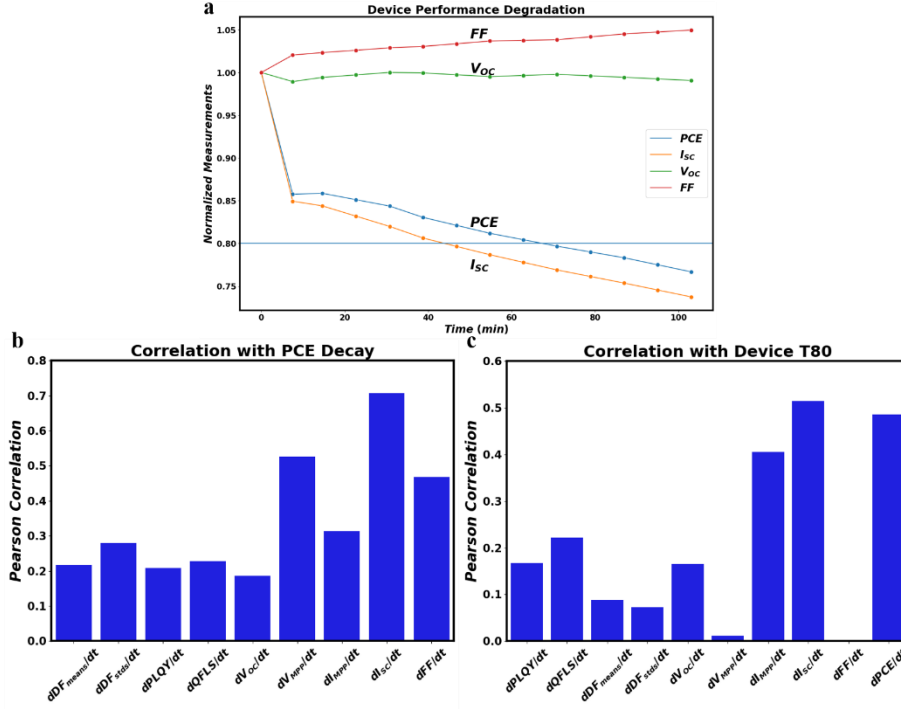


Figure 4 (a) Typical performance of a degrading device at 1 sun 25 °C 0RH 21% O₂. (b) Pearson correlation between the decay in PCE and the decay in other device measurements. (c) Pearson correlation between $T80$ and the decay of device measurements.

Testing the stability of a solar cell under realistic operating condition would be time-consuming and impractical in a laboratory setting. By increasing the stress intensity applied to the device, degradation can be accelerated, enabling data collection within a more reasonable timeframe. Figure 4a depicts the typical performance of a mixed Sn-Pb perovskite solar cell as it degrades. Our observation indicates that the loss in device PCE is mainly from the decay in the device's I_{sc} . In contrast, the V_{oc} and FF remains relatively stable, showing a slight increase over the course of the degradation period. In Figure 4b, our dataset reveals that the decay of I_{sc} has a Pearson correlation coefficient of 0.71 with the decay of PCE . It is important

to note that the decay of device performance was calculated from the initial 8 mins of degradation. Additionally, the prediction target $T80$ is highly correlated with the decay of I_{SC} and PCE showing a Pearson correlation of 0.51 and 0.49, respectively. These correlations suggest that the decay of I_{SC} is potentially an important features for predicting $T80$ in these devices.

3.3 Feature Correlation Analysis

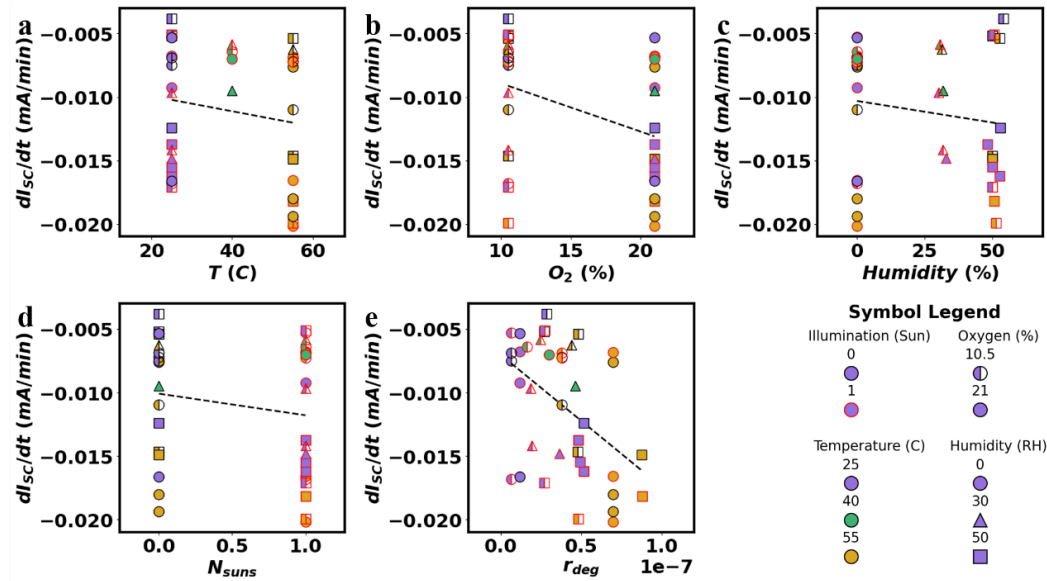


Figure 5 Distribution of collected degradation data of the decay in I_{SC} with (a) temperature, (b) O_2 concentration, (c) relative humidity, (d) illumination, and (e) degradation rate for perovskite.

The effect of different environmental factors such as temperature, oxygen, humidity and illumination on the decay of devices' performance was explored. Figure 5a, 5b, 5c, and 5d, show that higher levels of all environmental factors increased the decay of I_{SC} with oxygen having the most pronounced. This observation is consistent with previous studies where O_2 had the largest impact on the diffusion length of the perovskite,²¹ indicating that the reduction in I_{SC} is primarily due to a loss in diffusion

length. The degradation rate of the mix Sn-Pb perovskite material (r_{deg}), which was obtained from previous study on perovskite bare film²¹, is calculated with the following equation where k_B is the Boltzmann constant (8.617×10^{-5} eV/K), T is the temperature (K), P_{O_2} is the partial pressure of oxygen (kPa), and P_{H_2O} is the partial pressure of water (kPa):

$$r_{deg} = 0.19e^{\left(-\frac{0.5}{k_BT}\right)} \frac{P_{O_2}}{1 + 9.33 \times 10^{-3} \cdot P_{O_2}} + 5.18 \times 10^{-13} e^{\left(\frac{0.21}{k_BT}\right)} \frac{P_{O_2} P_{H_2O}}{1 + 9.33 \times 10^{-3} \cdot P_{O_2}} \quad (8)$$

In our study, r_{deg} shows the strongest correlation with the decay of I_{SC} , with a Pearson correlation of -0.47. This finding also aligns with the machine learning model used to predict $1/t_{LD,80}$ which identified the decomposition rate of the perovskite as the most dominant feature.²¹

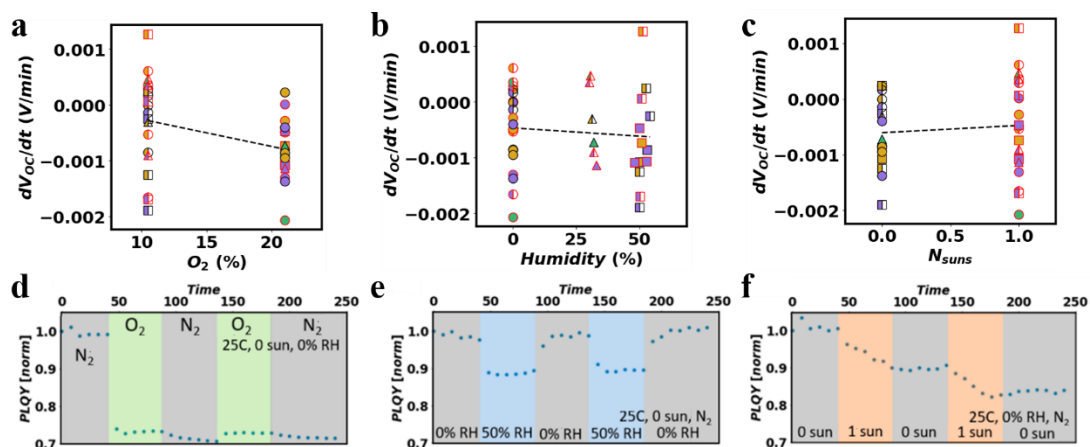


Figure 6 Distribution of collected degradation data of the decay in V_{OC} with (a) oxygen concentration, (b) relative humidity, (c) illumination. (Refer to Figure 5 for symbol legend.) Normalized photoluminescence quantum yield (PLQY) of perovskite bare film where (d) oxygen (e) humidity, and (f) illumination is added and removed when degrading the film. The units of time are minutes. Adapted with permission from Meng, Y.; Sunkari, P. P.; Meilā, M.; Hillhouse, H. W. Chemical Reaction Kinetics of the Decomposition of Low-Bandgap Tin-Lead Halide Perovskite Films

and the Effect on the Ambipolar Diffusion Length. ACS Energy Lett 2023, 8 (4), 1688–1696. Copyright 2024 American Chemical Society.

Although the V_{OC} typically remains stable with degrading devices, we investigated if environmental factors could potentially affect the decay of V_{OC} . Figure 6a, 6b, and 6c indicate that oxygen has the significant impact, causing a more substantial decrease in V_{OC} with higher oxygen concentration, while humidity and illumination showed minimal to no effect. This observation is consistent with previous studies on bare mixed Sn-Pb perovskite films where various environmental factors were introduced and removed to assess their impact on the film's photoluminescence quantum yield ($PLQY$),²¹ which correlates with device V_{OC} .³¹ In the study, the presence of oxygen drastically reduced the $PLQY$ where humidity and illumination caused only minor decreases during the initial 8 minutes of exposure. Indicating the formation of non-perovskite phases and deep defects in devices in the presence of oxygen.

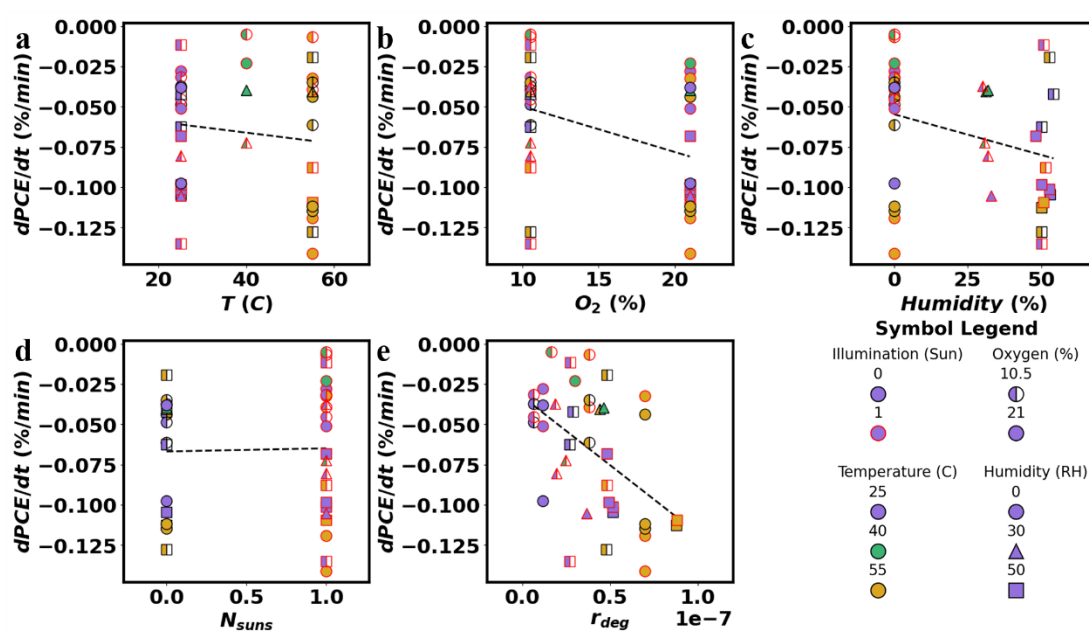


Figure 7 Distribution of collected degradation data of the decay in PCE with (a)

temperature, (b) O_2 concentration, (c) relative humidity, (d) illumination, and (e) degradation rate for perovskite.

The decay in I_{SC} and V_{OC} are further reflected in the decay of PCE where an increased correlation with oxygen and humidity is observed, as shown in Figure 7b, 6c. More importantly, the decay of PCE had the highest correlation with r_{deg} , indicating that it effectively captures the complex interaction of various environmental factors on device performance.

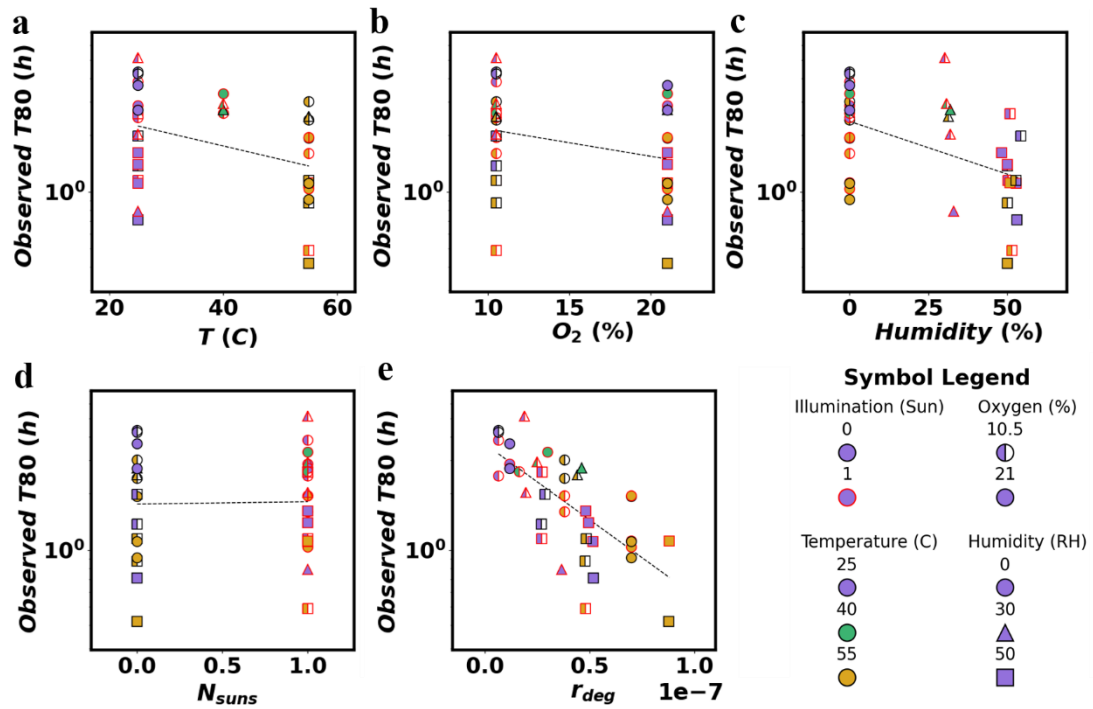


Figure 8 Distribution of collected degradation data of the decay in $T80$ with (a) temperature, (b) O_2 concentration, (c) relative humidity, (d) illumination, and (e) degradation rate for perovskite.

Studying the correlation of each environmental factor and r_{deg} with the prediction target of $T80$ reveals that higher humidity, O_2 concentration, and temperature contributed to the decrease in device lifetime shown in Figure 8a, 7b, and

7c. However, these environmental factors also result in a higher r_{deg} which exhibits the highest Pearson correlation of -0.71 with $T80$. Suggesting that the degradation rate expression to be the most significant predictor for $T80$, indicating that the rate at which the perovskite material degrades is the critical factor in forecasting the operational lifespan of the devices. The rate-based feature includes the effects of temperature (T), oxygen (P_{H_2O}), moisture (P_{H_2O}) and the illumination intensity (I_{in}), thereby encompassing the complex, non-linear relationships among the environmental stressors. This comprehensive modeling approach, which challenges the limitations of linear models that consider environmental stresses as independent features, highlights the efficacy of the rate expressions developed in preceding studies.

3.4 Machine Learning Model to Predict $T80$

The dataset used for $T80$ modeling consists of 39 runs ($N = 39$) representing 24 distinct environmental conditions. Features are calculated from data collected during degradation experiments and are categorized into three classes.

1. *a priori* known features: These variables are environmental factors such as temperature, humidity, oxygen concentration, and light intensity to which the device is subjected during degradation. Additionally, the kinetic rate expression for $FA_{0.75}Cs_{0.25}Pb_{0.5}Sn_{0.5}I_3$ degradation calculated from Eq (8) is also included.
2. Experimental features: These features are derived from initial measurements of the device parameters.
3. Time derivative features: These include the 1st and 2nd time derivative of the device's parameters, estimated from the first 8 minutes for the 1st time derivative and 16 minutes for the 2nd time derivative of each experiment. This

class include sample-specific measurements based on *PL* images, *DF* images, and *J–V* measurements made during the early cycles of data acquisition. Parameters include its quasi-Fermi level splitting (*QFLS*), *DF*, dark discharge (*Q_{dis}*), *V_{OC}*, *I_{SC}*, *FF*, and *PCE*.

This comprehensive feature set yields a total of 25 features summarized in Table 1. This structured approach ensures that the model captures the critical aspects of device degradation and enables accurate prediction of *T80*.

Table 1 Feature provided to machine learning models that predict perovskite solar cell *T80* and how they are obtained.

Feature Symbol	Physical interpretation/calculation method
<i>A priori</i> known features	
$1/T$	Inverse of solar cell temperature, controlled during experiment
p_{H_2O}	Partial pressure of H ₂ O, calculated from relative humidity measurement
p_{O_2}	Partial pressure of oxygen, calculated from oxygen percentage
N_{suns}	Number of suns illuminated on solar cell, controlled during experiment
r_{deg}	Decomposition rate of FA _{0.75} Cs _{0.25} Pb _{0.5} Sn _{0.5} I ₃ under specified environmental conditions, calculated from kinetic model described in Meng <i>et al</i> ¹⁷
Experimental features	
$QFLS (t = 0)$	Quasi-Fermi level splitting, calculated from device photoluminescence quantum yield (<i>PLQY</i>) at the start of the experiment
$DF_{mean} (t = 0)$	Mean dark field pixel intensity at the start of the experiment
$DF_{std} (t = 0)$	Standard deviation dark field pixel intensity at the start of the experiment
$V_{OC} (t = 0)$	Open-circuit voltage at the start of the experiment, taken from measurements at open circuit
$I_{SC} (t = 0)$	Short-circuit current at the start of the experiment, taken from measurements at short circuit
$FF (t = 0)$	Fill factor at the start of the experiment, taken from

	measurements at open circuit, short circuit, and maximum power point
$PCE(t = 0)$	Power conversion efficiency at the start of the experiment
$Q_{dis}(t = 0)$	Dark discharge measurements of the device at the start of the experiment
Time derivative features	
$dQFLS/dt$	1 st time derivative of $QFLS$, estimated from the first 8 minutes of each experiment
dDF_{mean}/dt	1 st time derivative of DF_{mean} , estimated from the first 8 minutes of each experiment
dDF_{std}/dt	1 st time derivative of DF_{std} , estimated from the first 8 minutes of each experiment
dV_{oc}/dt	1 st time derivative of V_{oc} , estimated from the first 8 minutes of each experiment
dI_{sc}/dt	1 st time derivative of I_{sc} , estimated from the first 8 minutes of each experiment
dFF/dt	1 st time derivative of FF , estimated from the first 8 minutes of each experiment
$dPCE/dt$	1 st time derivative of PCE , estimated from the first 8 minutes of each experiment
d^2QFLS/dt^2	2 nd time derivative of $QFLS$, estimated from the first 16 minutes of each experiment
d^2V_{oc}/dt^2	2 nd time derivative of V_{oc} , estimated from the first 16 minutes of each experiment
d^2I_{sc}/dt^2	2 nd time derivative of I_{sc} , estimated from the first 16 minutes of each experiment
d^2FF/dt^2	2 nd time derivative of FF , estimated from the first 16 minutes of each experiment
d^2PCE/dt^2	2 nd time derivative of PCE , estimated from the first 16 minutes of each experiment

Several popular feature selection methods for small dataset were used to see which feature would be selected more often indicating its importance in predicting device $T80$. Given the size of the dataset (39 runs), the large feature menu poses a risk of overfitting. To mitigate this risk, the feature menu was divided into three parts and fed into the machine learning model to assess the importance of different features. The models are evaluated using leave-one-out cross-validation (LOOCV), where each experiment in the dataset is sequentially left out as a test sample, and the remaining

data are used to train the model. Statistics from the distribution of testing errors provide an unbiased estimate of the model's predictive accuracy on unseen data, serving as its ultimate measure of performance. The stability of the models concerning the training set can be assessed by comparing the feature weights across iterations of the test/train split. Stable models will consistently select the same features and assign weights with similar magnitudes, while feature weights in unstable models may vary significantly.

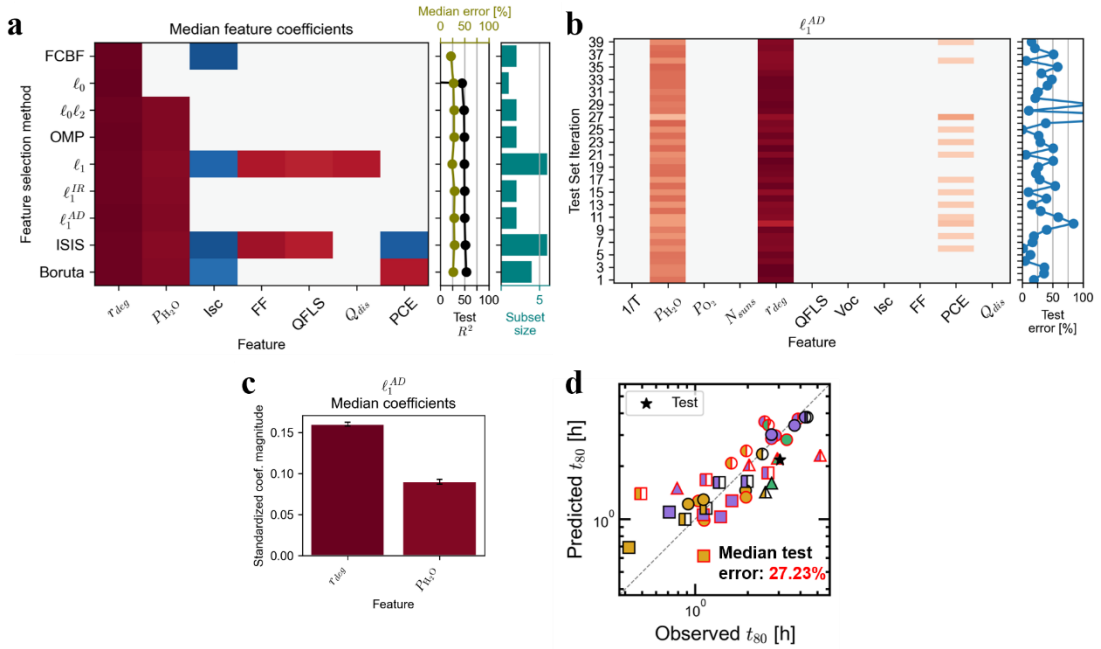


Figure 9 Modeling result with 13 features using multiple feature selection methods. (a) Heatmap showing the features are selected by each method. As well as the median error, test R^2 , and subset size on the right. (b) Coefficient values for each test/train split and its test error and a (c) bar plots of feature coefficients with ℓ_1^{AD} method. Positive and negative values are indicated by blue and red hues, deeper shades are of greater magnitude. (d) Parity plot of a LOOCV with a median test error of 27.23%.

Initially, only the environmental conditions and the initial performance of the device were utilized as features, resulting in a total of 13 features ($p = 13$). Among

all methods, the decomposition rate of $\text{FA}_{0.75}\text{Cs}_{0.25}\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_3$ (r_{deg}) emerged as the most dominant feature, as depicted in Figure 9a. Other significant features included the partial pressure of water (P_{H_2O}). The ℓ_1^{AD} method demonstrated high stability where the same features were selected in most iteration, as shown in Figure 9b. A simple linear model is constructed using the selected features where the magnitudes of their coefficients are illustrated in Figure 9c. The parity plot in Figure 9d shows the results of LOOCV performed on the model resulting in a median test error of 27.23%.

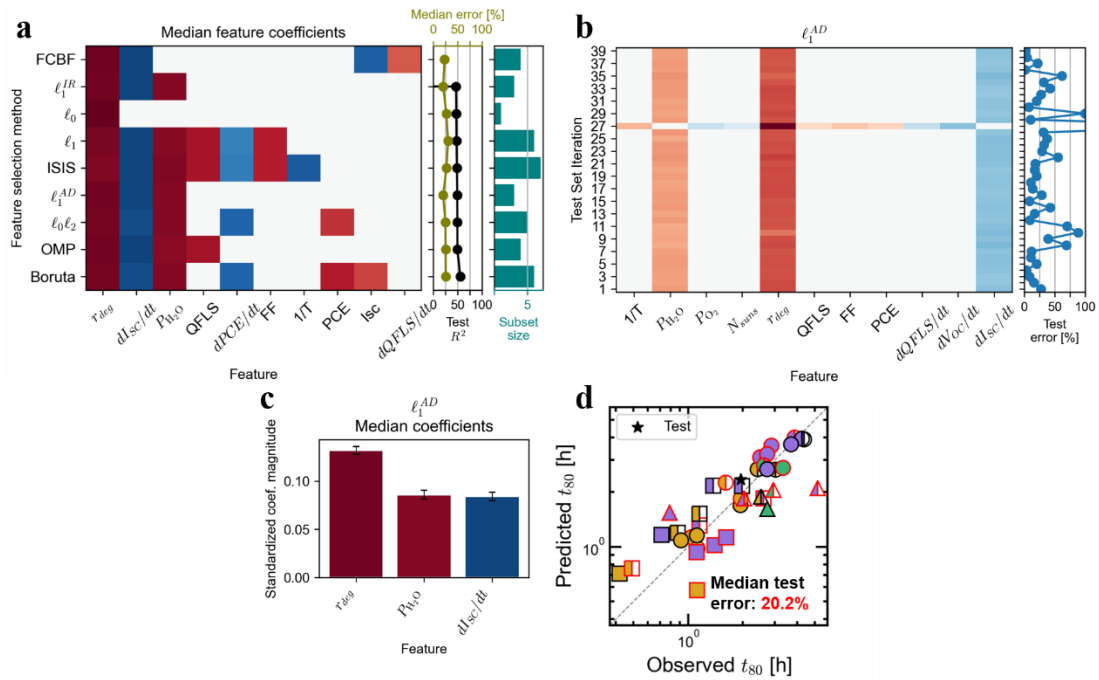


Figure 10 Modeling result with 20 features using multiple feature selection methods. (a) Heatmap showing the features are selected by each method. As well as the median error, test R^2 , and subset size on the right. (b) Coefficient values for each test/train split and its test error and a (c) bar plots of feature coefficients with ℓ_1^{AD} method. Positive and negative values are indicated by blue and red hues, deeper shades are of greater magnitude. (d) Parity plot of a LOOCV with a median test error of 20.2%.

Subsequently, we expanded the feature set to include the 1st time derivative of

the device experimental measurements, increasing the number of features to 20 ($p = 20$). Figure 10a reveals that r_{deg} and P_{H_2O} remain as dominant features across different feature selection methods. Additionally, newly introduced features, such as the slope of I_{SC} , was also selected. The ℓ_1^{AD} method exhibited high stability, consistently selecting the same features in most iterations, as depicted in Figure 10b. Although the inclusion of additional features introduced some degree of overfitting in several feature selection methods where 5 or more features were chosen, there is an improvement in the prediction accuracy of T_{80} , where we are able to obtain a median test error of 20.2% with a sparse feature model. This indicates that incorporating both *a priori* known features and initial decay of device parameter features are essential in providing an accurate prediction of T_{80} .

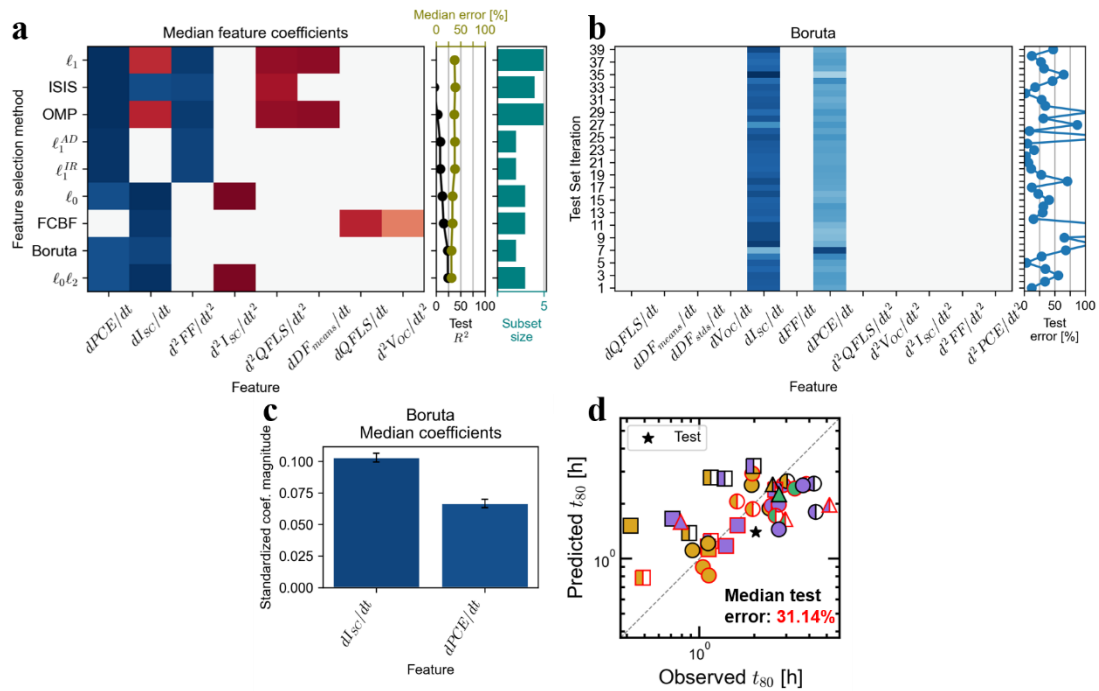


Figure 11 Modeling result with 12 features using multiple feature selection methods. (a) Heatmap showing the features are selected by each method. As well as the median error, test R^2 , and subset size on the right. (b) Coefficient values for each test/train split and its test error and a (c) bar plots of feature coefficients with *Boruta* method.

Positive and negative values are indicated by blue and red hues, deeper shades are of greater magnitude. (d) Parity plot of a LOOCV with a median test error of 31.14%.

Lastly, we conducted an analysis by removing all the *a priori* known features as well as the experimental features. Instead, we added the 2nd time derivative of the device experimental measurements, which is estimated from the first 16 minutes of each experiment, to explore whether it could capture any non-linearity in the decay of device performance. This resulted in a feature list with 12 features ($p = 12$). Figure 11a illustrates that the early time decay of I_{SC} and PCE were the most dominant features being selected by a majority of methods. The Boruta method exhibited high stability, selecting the same features throughout all iterations, as depicted in Figure 11b. The resulting model had a higher median test error of 31.14% from a LOOCV, as displayed in Figure 11d. This substantial decrease in accuracy reflects the importance of having *a priori* known features when predicting the $T80$ for solar cells.

4. Conclusions

In this study, we have compiled degradation data for ITO/PEDOT:PSS/FA_{0.75}Cs_{0.25}Pb_{0.5}Sn_{0.5}I₃/C₆₀/BCP/Ag solar cells under a range of environmental conditions. We discovered that decline in power conversion efficiency is primarily attributed to the deterioration in short-circuit current, while the fill factor and open-circuit voltage remain high over the effective lifetime of the devices. The main cause for the decay in short-circuit current is the presence of O₂ and H₂O which is associated with the formation of defects and loss of diffusion length. The kinetically modeled FA_{0.75}Cs_{0.25}Pb_{0.5}Sn_{0.5}I₃ decomposition rate exhibited the highest correlation with device lifetime ($T80$), a correlation that was further validated by our machine learning model trained to predict $T80$. Models developed using different feature sets

consistently incorporated sparse features, including the perovskite degradation rate, partial pressure of water, early time decay in short-circuit current, and early time decay of power conversion efficiency. These findings highlight the importance of leveraging the derived kinetic rate equation from prior research in constructing a robust machine learning model capable of predicting the longevity of low band-gap solar cells accurately.

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