## Supplementary Information: Bayesian Optimization and Prediction of the Durability of Triple-Halide Perovskite Thin Films under Light and Heat Stressors

Deniz N. Cakan,<sup>a</sup> Eric Oberholtz,<sup>a</sup> Ken Kaushal,<sup>a</sup> Sean P. Dunfield,<sup>a</sup> and David P. Fenning<sup>a</sup>

## Contents

## List of Tables

S1	Compositions of endpoint solutions used for interpolation within $FA_{0.78}Cs_{0.22}Pb(I_xBr_yCl_z)_3$ .	2
List of	Figures	
S1	LED spectrum used in durability testing	3
S2	Model comparison	4
S3	X-ray-diffraction and scanning electron microscopy	5
S4	Voltage deficit of $FA_{0.78}Cs_{0.22}Pb(I_xBr_yCl_z)_3$ films	6
S5	PLQY of $FA_{0.78}Cs_{0.22}Pb(I_xBr_yCl_z)_3$ films	7
S6	Devices from initial search	8
S7	Pearson's correlation matrix of half-cells	9
S8	Raw feature values	10
S9	Histogram of coefficient of variation RMSE	11
S10	Best performing RFR	12
S11	Pearson's correlation matrix of films	13
S12	RFR learning curves	14

<sup>&</sup>lt;sup>0a</sup> Aiiso Yufeng Li Family Department of Chemical and Nano Engineering, University of California San Diego, La Jolla, CA, 92093, USA; E-mail: dfenning@ucsd.edu

**Table S1** Compositions of endpoint solutions used for interpolation within  $FA_{0.78}Cs_{0.22}Pb(I_xBr_yCl_z)_3$ .

$I_X$	$Br_y$	$Cl_z$
0.95	0.05	0
0.95	0	0.05
0.80	0.20	0
0.80	0.15	0.05



Figure S1 LED spectrum used for the 1-sun test.



**Figure S2** GPR negative absolute error comparison between model produced by samples produced via grid search vs samples produced in BO with EI at 100% exploitation. Color bar yellow depicts regions of relatively minimal difference.



**Figure S3** a) XRD and b-d) SEM of  $FA_{0.78}Cs_{0.22}Pb(I_xBr_yCl_z)_3$  films, label inset with Br fraction. Scale bar  $2\mu$ m.



**Figure S4** Voltage deficit of  $FA_{0.78}Cs_{0.22}Pb(I_xBr_yCl_z)_3$  films.



Figure S5 PLQY of  $FA_{0.78}Cs_{0.22}Pb(I_xBr_yCl_z)_3$  films.



**Figure S6** Devices from initial search containing 15 composition unique  $FA_{0.78}Cs_{0.22}Pb(I_xBr_yCl_z)_3$  films. a) Schematic of device architecture used in this experiment. b) Photograph of completed 1 cm<sup>2</sup> devices in the automatic JV tester system. c) JV curves of the 15 unique samples (3 repeats). d) Box plot with overlaid violin plot of the PCE distribution.

												- 1.00	
PL_intensity -		0.21	0.45	-0.14	-0.11	-0.18	-0.15	0.3	-0.48	0.71			
PL_peakenergy -	0.21		0.61	0.037	0.1	0.0086	0.0095	-0.23	0.17	0.065		- 0.75	
PL_fwhm -	0.45	0.61		-0.087	-0.015	-0.039	-0.073	-0.18	0.14	0.032		- 0.50	
PCE -	-0.14	0.037	-0.087					-0.18	0.2	-0.16		- 0.25	ation
FF -	-0.11	0.1	-0.015					-0.32	0.32	-0.15		- 0.00	Correla
Voc -	-0.18	0.0086	-0.039					-0.56	0.54	-0.21			son's (
Jsc -	-0.15	0.0095	-0.073					0.063	-0.0067	-0.14		0.25	Pear
-	0.3	-0.23	-0.18	-0.18	-0.32	-0.56	0.063					0.50	
Br -	-0.48	0.17	0.14	0.2	0.32	0.54	-0.0067					0.75	
Cl -	0.71	0.065	0.032	-0.16	-0.15	-0.21	-0.14					- 1.00	
	PL_intensity -	PL_peakenergy -	- PL_fwhm	PCE -	Ŧ	Voc -	Jsc -	_	Br -	Ū		1.00	

**Figure S7** Calculated Pearson's correlation matrix between PL shift, FWHM, and intensity of PL spectra alongside device efficiency (PCE), fill factor (FF), open-circuit voltage (Voc), and short-circuit current density (Jsc) among ITO/MeO-2PACz/PSK/C60/BCP/Ag devices across the PSK compositional space.



**Figure S8** Raw values for characterization used in RFR model training. a) PL peak energy. b) PL peak intensity. c) Photodegradation PL peak shift. d)  $\Delta K$  extracted from ISOS-L2 testing. Regions of no experiment is shaded out. Color bar located below ternaries. e) PL full-width half max. f) Reverse scan PCE of half-cells.



Figure S9 Histogram of coefficient of variation RMSE.



Figure S10 a) Best RFR parity plot and b) corresponding Shap table.

![](_page_12_Figure_0.jpeg)

Figure S11 Calculated Pearson's correlation matrix between input variables used in the random forest regression.

![](_page_13_Figure_0.jpeg)

Figure S12 Learning curves showing model's a) R<sup>2</sup> and b) -MSE, as a function of training size.