Supplementary Information (SI) for Sustainable Energy & Fuels.

Optimizing Lead-Free Cs2AgBiBr6 Double Perovskite Solar Cells: A Comprehensive Study Using Realistic Simulation Approach

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SCAPS Simulation Technique

The SCAPS software functions as a one-dimensional simulation tool that can model up to seven semiconductor layers, accounting for interface and error analysis within each layer. The software performs calculations of various parameters such as current-voltage (I-V) characteristics, energy bands, current density-voltage (J-V) characteristics, photovoltaic (PV) parameters, energy band diagrams, particle concentration, alternating current, and graphs will generate (AC) characteristics (measurement of capacitance (C) and conductivity (G) versus voltage (V) and frequency (f)) and spectral response (quantum efficiency, QE). These analyzes are based on the core semiconductor equations, called Equations 4-6, which include the continuity equations for electrons and holes, and the Poisson equation.

$$\frac{D}{dx}\left(-\varepsilon(x)\frac{d\phi}{dx}\right) = [p(x) - n(x) + ND(x) - NA(x) + p(x) - n(x)] \quad \text{(S1)}$$

here ND represent the donor density and acceptor density is represented by NA, and p represents hole density. The density of trapped electrons is expressed by n(x) and holes is expressed in terms n(p), and the medium's dielectric constant is expressed as ε. The one-dimensional continuity equation for electrons and holes is expressed as follows.

$$\frac{\partial n(x,t)}{\partial t} = \frac{1}{q}\frac{\partial J_n}{\partial x} + Gn(x,t) - \mathrm{Rn}\,(x,t) \quad \text{(S2)}$$

$$\frac{\partial p(x,t)}{\partial t} = -\frac{1}{q}\frac{\partial J_p}{\partial x} + Gp(x,t) - Rp(x,t) \quad \text{(S3)}$$

Electron density cm^{-3} (n), hole density cm^{-3} (p), charge (q), which is correlated with the carrier concentration of charge, and the recombination and production rates (R and G) are denoted in this framework. The drift current densities Jp represent the concentrations of holes and Jn the concentrations of electrons.

Architecture of the Devices & Simulation Parameters

Table S1: Parameters used for simulation study

Parameters	WO_3	$Cs_2AgBiBr_6$	NiO
Thickness ((nm)	150	500	50
Bandgap (eV)	3.15	1.7	3.8
Electron Affinity (eV)	4.55	4.19	1.8
Dielectric Permittivity, ε (relative)	10	5.8	10
CB Effective Density of States, Nc (1/cm3)	4.20 E18	1 E19	2.80 E18
VB Effective Density of States, Nv (1/cm3)	9.00 E18	11 E19	11 E19
Thermal Velocity of Electrons (cm/s)	1 E7	1 E7	1 E7
Thermal Velocity of Holes (cm/s)	1 E7	1 E7	1 E7
Mobility of Electrons, µn (cm2 /Vs)	20	11.81	12
Mobility of Holes, µp (cm2 /Vs)	10	0.49	2.8
Donor-Like Doping Concentrations, ND (1/cm3)	1E19	0	0
Acceptor-Like Doping Concentrations, NA (1/cm3)	0	1E16	1E19
Defect Density, Nt (1/cm3)	1E14	1E14	1E14

FDTD Simulation

In fig S1, our simulation is structured around a standard geometry consisting of four distinct layers centered around a perovskite structure. At the core is a 600nm thick perovskite layer, flanked by a 50nm HTL and a 50nm ETL. The structure is topped with a 150nm FTO layer similar to that of the SCAPS 1D simulation. Employing the refractive index and extinction coefficient as a fundamental optical property, the solar cell's performance was assessed using FDTD simulations.

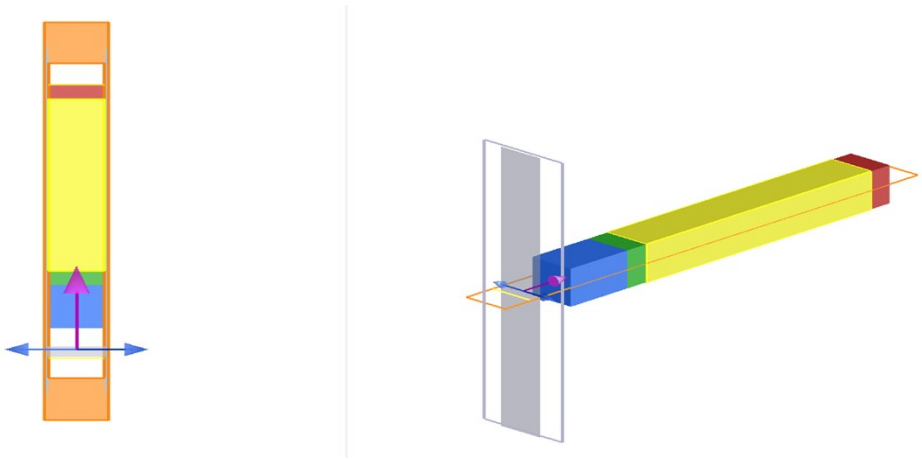

Fig S1: FDTD simulation structure

FDTD simulations are conducted in 2D/3D space. Here we conducted simulation on the PSC structure using mesh cells with an auto non-uniform mesh. The source of light in the simulation was set up as a plane wave mode along the y-axis, with wavelength spectrum of 350 - 1100 nm and an amplitude of 1. Perfect matched layers (PML) were applied in the y-axis to optimize incident light trapping, while periodic boundary conditions (BC) were applied along the x-direction to simulate infinite periodicity of the structure. To measure absorption, monitor of frequency-domain field and power have been placed adjacent to the layer under investigation, with additional monitors positioned at the front and back of the model for a comprehensive evaluation. In this study, the light absorption ratio (A_{abs}) is determined using an equation that incorporates the reflectance R(λ) and transmittance T(λ) of cell's front and rear surfaces. These two parameters, T(λ) and R(λ), are calculated using the FDTD method to evaluate the solar cell's light handling characteristics.

$$A_{abs}(\lambda) = 1 - R(\lambda) - T(\lambda) \qquad \text{(S4)}$$

Impact on internal quantum efficiency (IQE)

Current density–voltage (J–V) characteristic of a solar cell is typically governed by the diode law. Based on the diode equation, the V_{OC} of a single-junction solar cell can be described as:

$$V_{OC} = \frac{nkT}{q} ln\left(\frac{J_{SC}}{J_0} + 1\right) \quad \text{(S5)}$$

where n is the ideality factor, q is the elementary charge, k is Boltzmann constant, T is the absolute temperature, and J_{SC} and J_0 are the short - circuit and reverse saturation current densities, respectively.[1] We got Jsc value from FDTD simulation for all different thickness of absorber layer. To find the reverse saturation current densities, we used the following equation:

$$J_0(T, E_G(300K)) = A^* \cdot T^3 \cdot e^{-E_G(300K)/kT} \quad \text{(S5)}$$

Here, A^* is the Richardson constant, which is a measure of the number of states per unit energy, T is the temperature in Kelvin, $EG(300K)$ is the energy gap at 300 K, which is the energy difference between the valence band and the conduction band and k is the Boltzmann's constant.

The quantum efficiency of a solar cell, QE (λ), is defined by the following equation:

$$QE(\lambda) = \frac{P_{abs}(\lambda)}{P_{in}(\lambda)} \quad \text{(S6)}$$

where P_{in} (λ) and P_{abs} (λ) are the powers of the incident light and absorbed light within the solar cell, respectively, at a wavelength λ.

[1] C. Li *et al.*, "Reducing Saturation-Current Density to Realize High-Efficiency Low-Bandgap Mixed Tin–Lead Halide Perovskite Solar Cells," *Advanced Energy Materials,* vol. 9, no. 3, p. 1803135, 2019, doi: https://doi.org/10.1002/aenm.201803135.