Supplementary Materials

27.20% efficiency for lead-free double perovskite solar cell with all inorganic Cs_2BiAgI_6 using AZO UTL as a passivation layer

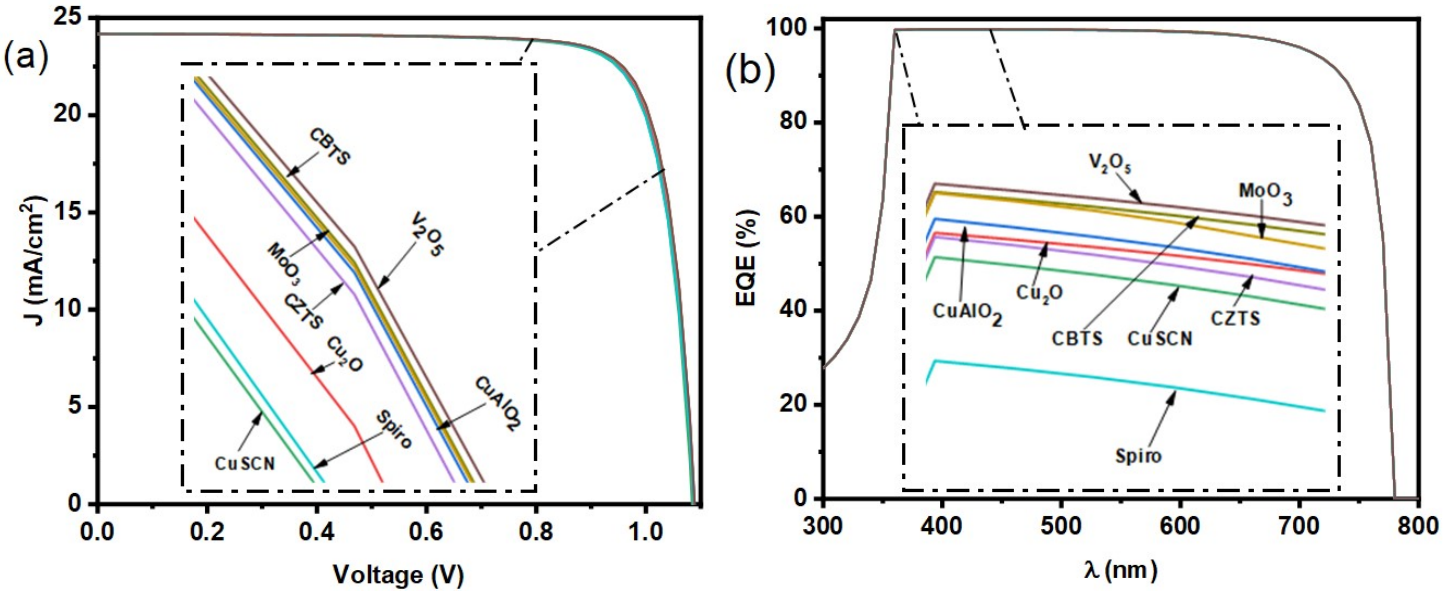
Aminreza Mohandes, Mahmood Moradi, *1

Department of Physics, College of Science, Shiraz University, 71946-84795 Shiraz, Iran. E-mail: mmoradi@shirazu.ac.ir

CONTENT

Table S1 PV output parameters of 16 devices for two ETLs and eight HTLs material of Cs_2BiAgI_6-DPSC, ITO/ETLs/Cs_2BiAgI_6 (800 nm)/HTLs/Au

No.	Device before optimization	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)
1	ITO/ZnO/AZO/Cs_2BiAgI_6/CBTS/Au	1.0893	24.18	81.89	21.57
2	ITO/ZnO/AZO/Cs_2BiAgI_6/Cu_2O/Au	1.0864	24.17	81.62	21.44
3	ITO/ZnO/AZO/Cs_2BiAgI_6/$CuAlO_2$/Au	1.0899	24.17	81.84	21.56
4	ITO/ZnO/AZO/Cs_2BiAgI_6/CuScN/Au	1.0847	24.16	81.43	21.35
5	ITO/ZnO/AZO/Cs_2BiAgI_6/CZTS/Au	1.0883	24.17	81.90	21.55
6	ITO/ZnO/AZO/Cs_2BiAgI_6/MoO_3/Au	1.0894	24.17	81.90	21.57
7	ITO/ZnO/AZO/Cs_2BiAgI_6/Spiro/Au	1.0887	24.17	81.21	21.37
8	ITO/ZnO/AZO/Cs_2BiAgI_6/V_2O_5/Au	1.0897	24.17	81.93	21.59
9	ITO/ZnO/Cs_2BiAgI_6/CBTS/Au	1.0890	24.18	81.87	21.56
10	ITO/ZnO/Cs_2BiAgI_6/Cu_2O/Au	1.0862	24.17	81.60	21.43
11	ITO/ZnO/Cs_2BiAgI_6/$CuAlO_2$/Au	1.0896	24.17	81.82	21.55
12	ITO/ZnO/Cs_2BiAgI_6/CuScN/Au	1.0845	24.17	81.41	21.34
13	ITO/ZnO/Cs_2BiAgI_6/CZTS/Au	1.0881	24.17	81.88	21.54
14	ITO/ZnO/Cs_2BiAgI_6/MoO_3/Au	1.0891	24.17	81.87	21.56
15	ITO/ZnO/Cs_2BiAgI_6/Spiro/Au	1.0884	24.17	81.19	21.36
16	ITO/ZnO/Cs_2BiAgI_6/V_2O_5/Au	1.0894	24.17	81.91	21.58



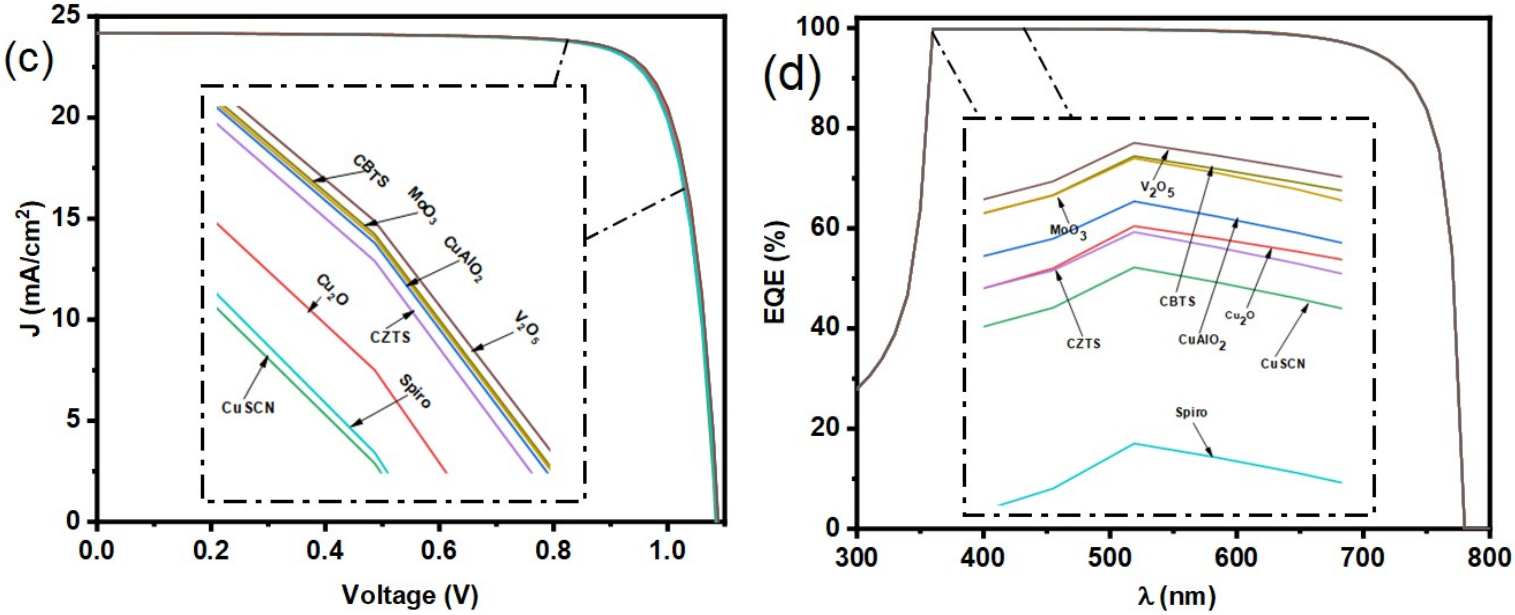
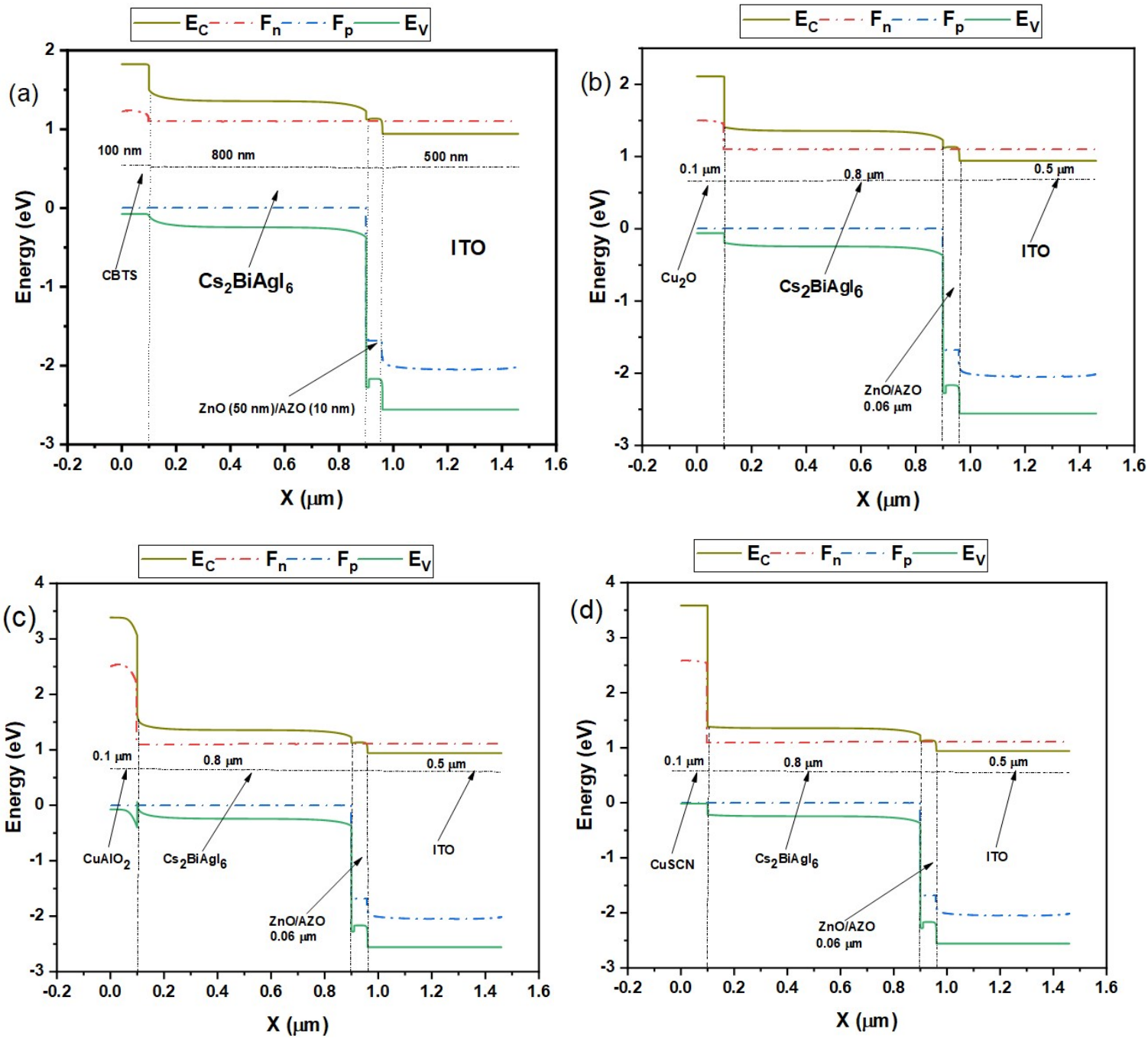
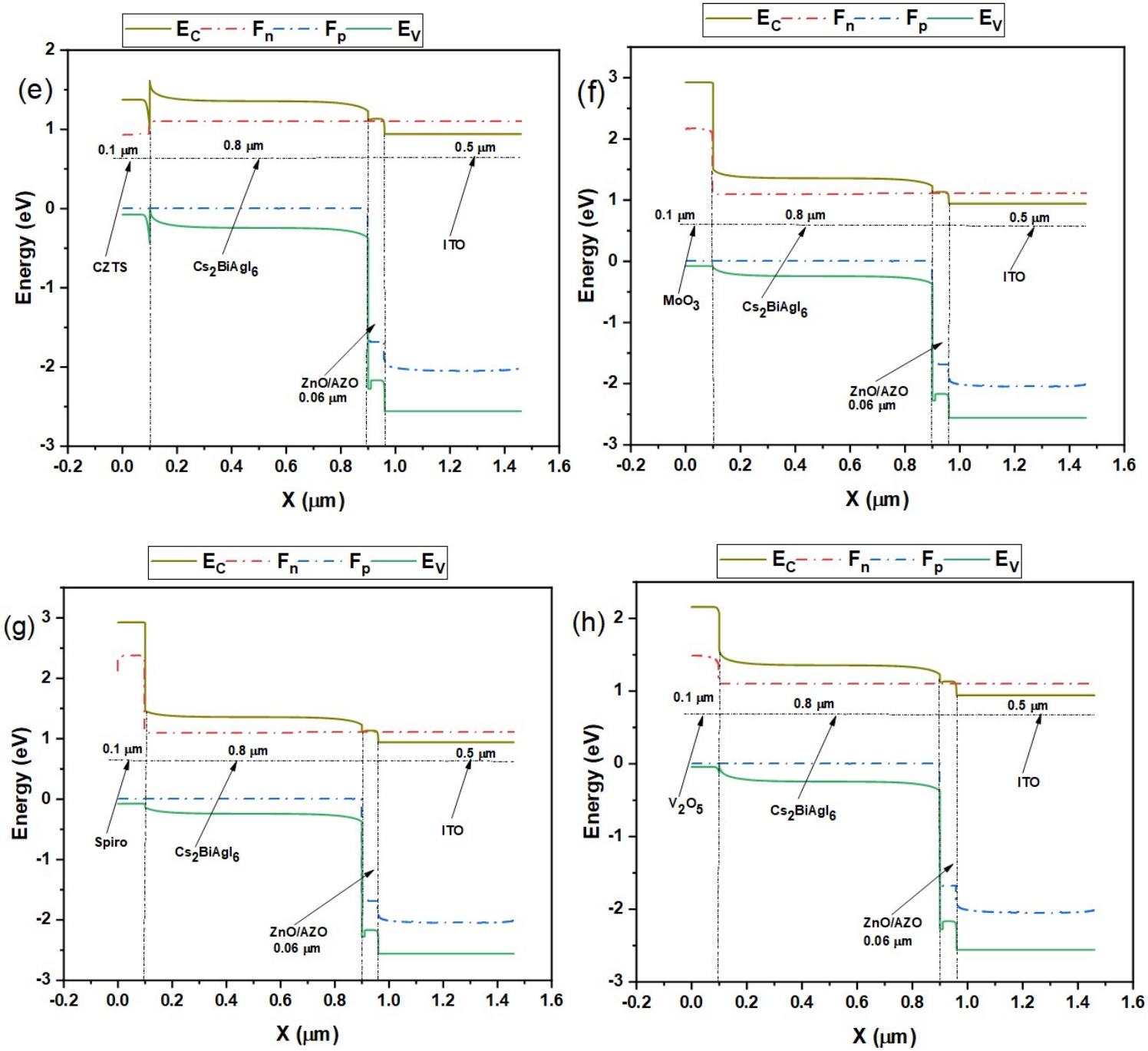
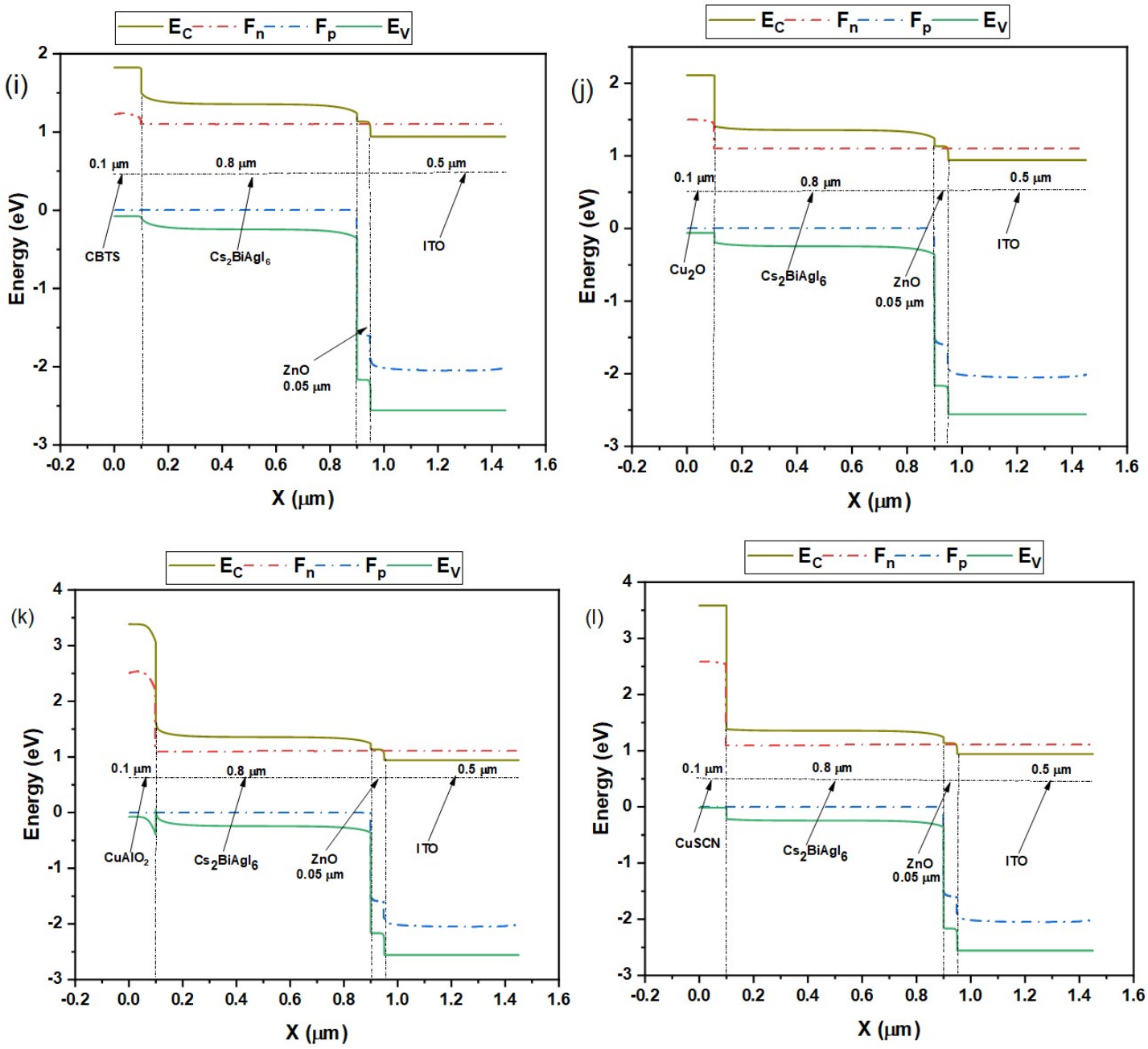


Fig. S1 The current density-voltage (J-V) and external quantum efficiency (EQE) curves (a and b) ZnO/AZO, and (c and d) ZnO. The eight HTLs are shown in the inset of the figures.


(a)
E_C F_n F_p E_V
100 nm
800 nm
500 nm
CBTS
Cs_2BiAgI_6
ITO
ZnO (50 nm)/AZO (10 nm)
Energy (eV)
X (μm)
(b)
E_C F_n F_p E_V
0.1 μm
0.8 μm
0.5 μm
Cu_2O
Cs_2BiAgI_6
ITO
ZnO/AZO
0.06 μm
Energy (eV)
X (μm)
(c)
E_C F_n F_p E_V
0.1 μm
0.8 μm
0.5 μm
$CuAlO_2$
Cs_2BiAgI_6
ITO
ZnO/AZO
0.06 μm
Energy (eV)
X (μm)
(d)
E_C F_n F_p E_V
0.1 μm
0.8 μm
0.5 μm
CuSCN
Cs_2BiAgI_6
ITO
ZnO/AZO
0.06 μm
Energy (eV)
X (μm)


(e)
(f)
(g)
(h)
E_C F_n F_p E_V
Energy (eV)
X (μm)
0.1 μm
0.8 μm
0.5 μm
CZTS
Cs_2BiAgI_6
ITO
ZnO/AZO
0.06 μm
MoO_3
Spiro
V_2O_5


(i)
(j)
(k)
(l)
E_C F_n F_p E_V
Energy (eV)
X (μm)
0.1 μm
0.8 μm
0.5 μm
CBTS
Cs_2BiAgI_6
ITO
ZnO
0.05 μm
Cu_2O
$CuAlO_2$
CuSCN

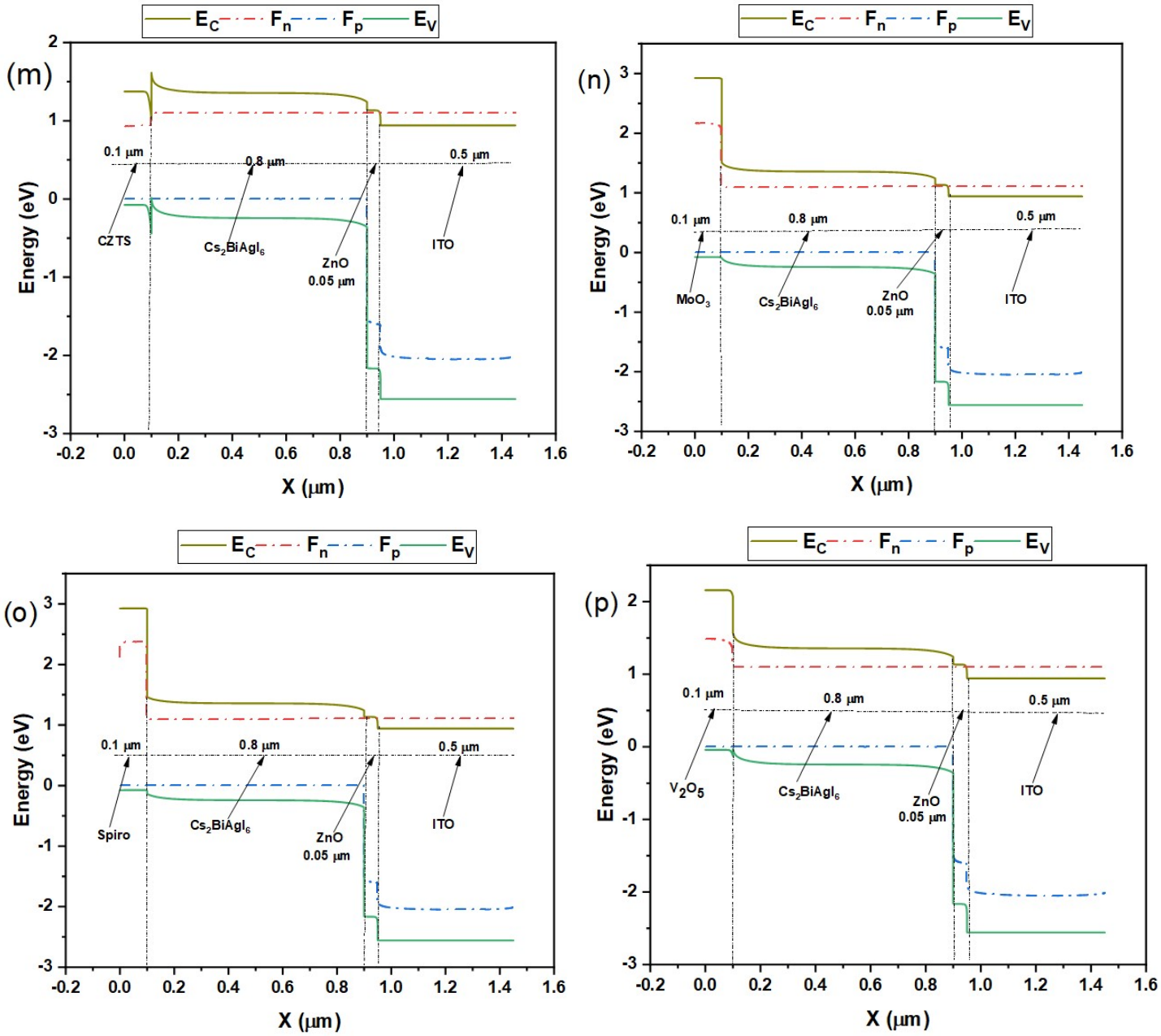


Fig. S2 Energy band alignment of Cs_2BiAgI_6-DPSC (a-h) ZnO/AZO, and (i-p) ZnO. (E_C (conduction band energy), F_n (quasi-fermi level of electron energy), F_p (quasi-fermi level of hole energy), and E_V (valence band energy)).

Table S2 Interfacial defect density parameters of ITO/ZnO/AZO/Cs_2BiAgI_6 (800 nm)/V_2O_5/Au device

Parameter	ZnO/AZO	AZO/Cs_2BiAgI_6	Cs_2BiAgI_6/V_2O_5
Defect type	Neutral	Neutral	Neutral
σ_n (cm^2)	1×10^{-17}	1×10^{-17}	1×10^{-18}
σ_p (cm^2)	1×10^{-18}	1×10^{-18}	1×10^{-19}
Distribution of energy	Single	Single	Single
$E_t - E_V$	Above the VB maximum	Above the VB maximum	Above the VB maximum
Energy level w.r.t. Reference (eV)	0.6	0.6	0.6
N_t (cm^{-2})	1×10^{10}	1×10^{10}	1×10^{10}

Table S3 PV output parameters of simulated Cs_2BiAgI_6-DPSC with variation of VBO_1 values, for ITO (500 nm)/ZnO (50 nm)/AZO (10 nm)/Cs_2BiAgI_6 (650 nm)/V_2O_5 (100 nm)/Au device

Band gap energy of HTL (eV)	VBO_1 (eV)	Structure type	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
1.5	-0.6	Cliff	1.0491	24.40	78.32	20.05
1.6	-0.5	Cliff	1.1462	23.79	79.48	21.68
1.7	-0.4	Cliff	1.2347	23.71	81.29	23.80
1.8	-0.3	Cliff	1.2946	23.85	82.15	25.98
1.9	-0.2	Cliff	1.3146	23.86	86.07	27
2	-0.1	Cliff	1.3193	23.84	86.36	27.17
2.1	**0.0**	**Cliff**	**1.3210**	**23.84**	**86.32**	**27.19**
2.2	+0.1	Spike	1.3213	23.83	86.31	27.18
2.3	+0.2	Spike	1.3213	23.83	86.31	27.18
2.4	+0.3	Spike	1.3213	23.83	86.31	27.18
2.5	+0.4	Spike	1.3214	23.83	86.29	27.17

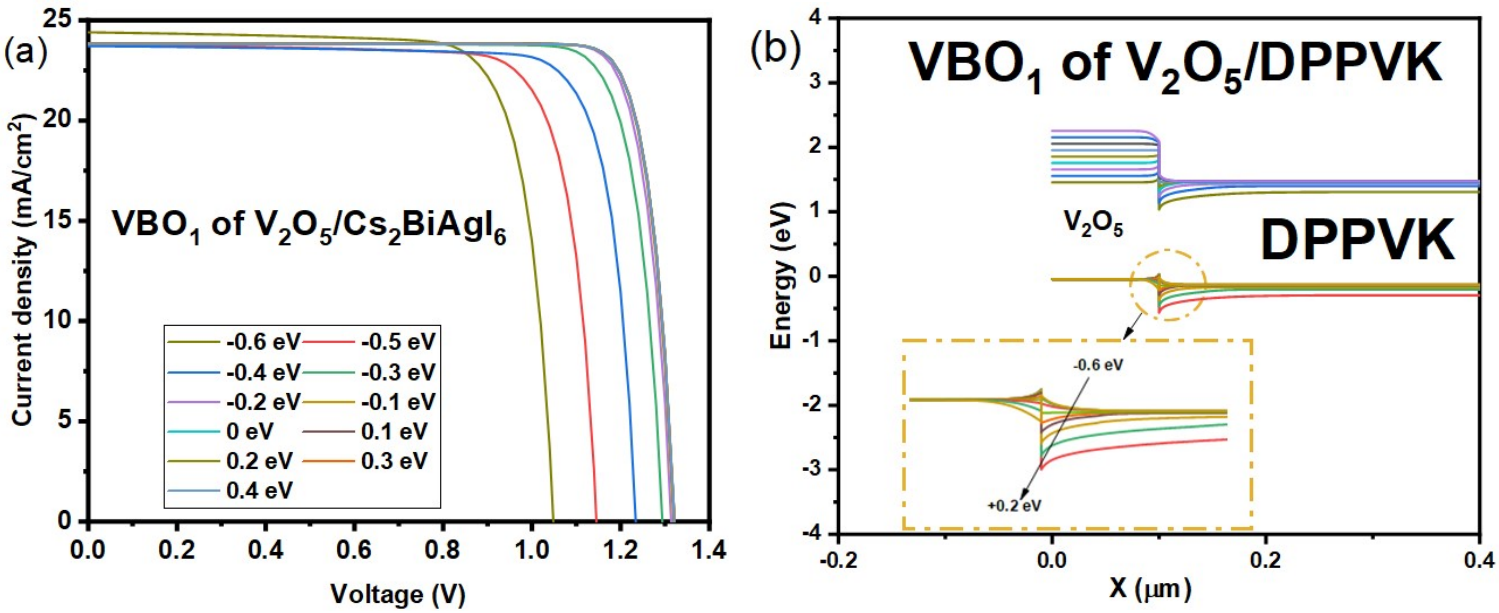


Fig. S3 (a) J-V curves, (b) energy band diagrams (EBDs), inset of figures demonstrate the variation of energy level of V_2O_5-HTL versus VBO_1 values.

Table S4 PV output parameters of simulated Cs_2BiAgI_6-DPSC with variation of CBO_1 values, for ITO (500 nm)/ZnO (50 nm)/AZO (10 nm)/Cs_2BiAgI_6 (650 nm)/V_2O_5 (100 nm)/Au device

Electron affinity of V_2O_5	CBO_1	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
3	0.4	1.2370	23.62	81.11	23.70
3.1	0.3	1.2951	23.80	84.11	25.93
3.2	0.2	1.3146	23.83	86.07	26.97
3.3	0.1	1.3193	23.84	86.36	27.16
3.4	**0.0**	**1.3210**	**23.84**	**86.32**	**27.19**
3.5	-0.1	1.3213	23.84	86.31	27.19
3.6	-0.2	1.3213	23.84	86.31	27.19

Table S5 PV output parameters of simulated Cs_2BiAgI_6-DPSC with variation of VBO_2 values, for ITO (500 nm)/ZnO (50 nm)/AZO (10 nm)/Cs_2BiAgI_6 (650 nm)/V_2O_5 (100 nm)/Au device

Energy band gap of AZO	VBO_2	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
3	-1.5	1.321	23.84	86.32	27.19
3.1	-1.6	1.321	23.84	86.32	27.19
3.2	-1.7	1.321	23.84	86.32	27.19
3.3	-1.8	1.321	23.84	86.32	27.19
3.4	**-1.9**	**1.321**	**23.84**	**86.32**	**27.19**
3.5	-2.0	1.321	23.81	86.30	27.14

Table S6 PV output parameters of simulated Cs_2BiAgI_6-DPSC with variation of CBO_2 values, for ITO (500 nm)/ZnO (50 nm)/AZO (10 nm)/Cs_2BiAgI_6 (650 nm)/V_2O_5 (100 nm)/Au device

Electron affinity of AZO (eV)	CBO_2 (eV)	Structure Type	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
3.5	+0.4	Spike	1.323	23.84	82.10	25.90
3.6	+0.3	Spike	1.3222	23.84	86.15	27.16
3.7	+0.2	Spike	1.3222	23.84	86.28	27.20
3.8	+0.1	Spike	1.3222	23.84	86.28	27.20
3.9	**+0.0**	**Cliff**	**1.3221**	**23.84**	**86.29**	**27.20**
4	-0.1	Cliff	1.321	23.84	86.32	27.19
4.1	-0.2	Cliff	1.3097	23.83	86.94	27.14

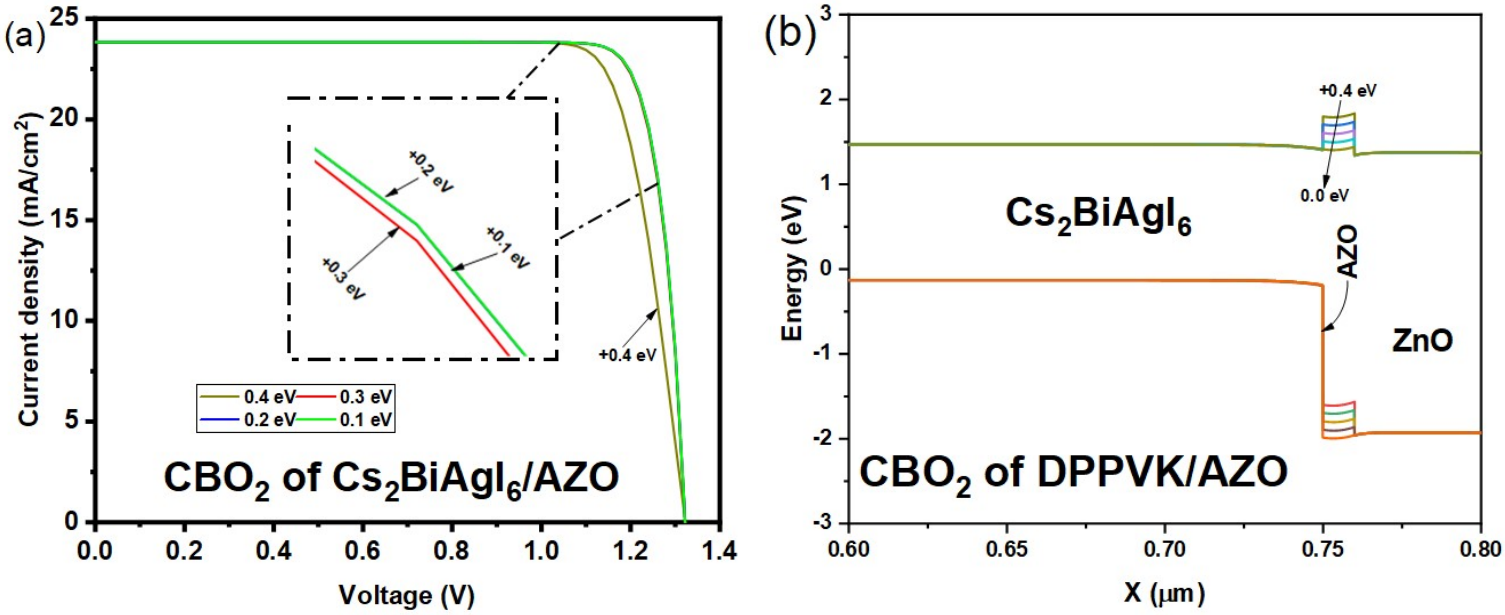


Fig. S4 (a) J-V curves, inset shows affinity variation, (b) energy band diagrams (EBDs) of AZO UTL for different values of CBO_2.

Table S7 Comparison of initial and final optimization of PV output parameters of the simulated device for ITO (500 nm)/ZnO (50 nm)/ AZO (10 nm)/Cs_2AgBiI_6 (650 nm)/V_2O_5 (100 nm)/Se device of Cs_2AgBiI_6-DPSC

No.	Parameters of Cs_2AgBiI_6-DPSC	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
1	Initial parameters (without optimization)	1.1005	23.78	82.78	21.67
2	Doping and Defect density of all layers optimization	1.3212	23.83	86.31	27.17
3	Energy level optimization	1.3221	23.84	86.28	27.20
4	Interfacial defect density optimization (final optimization)	1.3221	23.84	86.28	27.20

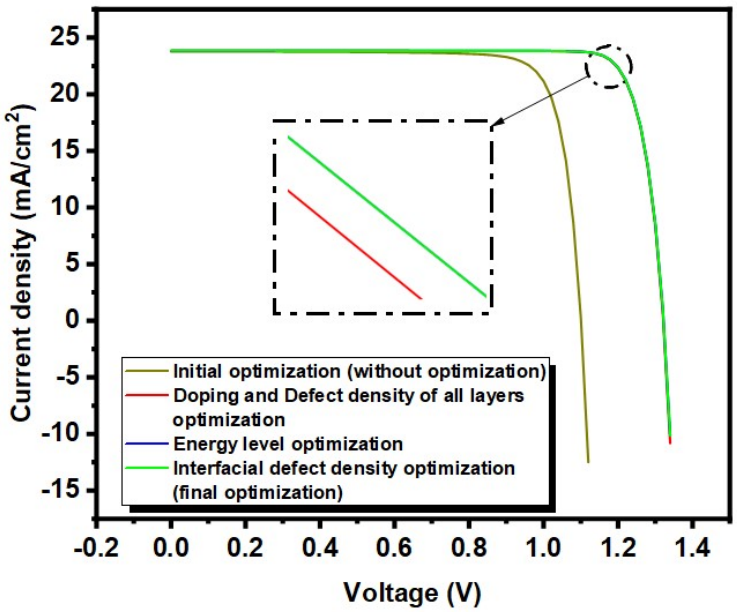


Fig. S5 J-V curve of the simulated DPSC, ITO (500 nm)/ZnO (50 nm)/AZO (10 nm)/Cs_2AgBiI_6 (650 nm)/V_2O_5 (100 nm)/Se device of Cs_2AgBiI_6-DPSC, illustrates the initial results and the final results after using the optimized parameters of doping and defect density, energy level, and interfacial defect density.