

**MONOLITHIC PEROVSKITE/ORGANIC TANDEM SOLAR CELLS WITH
IMIDAZOLE-BASED ORGANIC ADDITIVE**

TECHNICAL FIELD

[0001] The present disclosure generally relates to a solar cell for converting energy of light into electricity. In particular, the present disclosure relates to a solar cell having a perovskite-type active layer comprising 2-amino-4,5-imidazoledicarbonitrile (AIDCN) as an organic additive.

BACKGROUND

[0002] The Shockley-Queisser (S-Q) limit for single-junction solar cells has been an obstacle for improving power conversion efficiency (PCE) of emerging photovoltaics. Therefore, tandem solar cells integrating more than one kind of active layers are designed to bypass the above limit for higher power conversion efficiencies. Metal halide perovskite, with excellent semiconductor and material properties (such as long carrier lifetime, defect tolerance and availability of solution-processing, etc.) as well as broad bandgap tunability (about 1.2 to 3.0 eV), are ideal candidates for tandem applications. Despite the bandgap of perovskites being able to be easily tuned through manipulation of the ratio of halide anions (Br/I ratio in most cases), the incorporation of more than 20% bromide component for reaching a wider bandgap suitable for tandem solar cells can bring about halide phase segregation problem, which can adversely affect the efficiency and stability of the solar cells.

[0003] Halide phase segregation refers to a phenomenon that when exposed to continuous illumination, a wide-bandgap perovskite, with a bandgap greater than 1.8 eV and containing more than 20% bromide, will tend to form a segregated phase with lower Br/I ratio and lower bandgap as compared to the original state (*Hoke et al., Reversible photo-induced trap formation in mixed-halide hybrid perovskites for photovoltaics, Chem. Sci. (2015), 6, 613-617*).

Subsequent works reported that although the segregated phase only takes up about 1% volume, it will lead to a drastic carrier funneling process from the original phase to itself, thus seriously reduces the open-circuit voltage (V_{OC}) and creating stability issues of the devices.

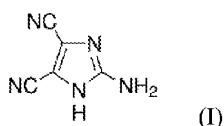
[0004] Several options are available for using wide-bandgap perovskite with a bandgap greater than 1.8 eV to form tandem solar cells. Commercialized silicon solar cell can be one option, but the high fabrication cost and difficulty in fabrication make it non-ideal in terms of solution-processing compatibility. Narrow bandgap perovskite containing Sn(II) was also reported to serve as another active layer, but the fast oxidation of Sn(II) to Sn(IV) brings problem for long-term stability. Comparing to the above options, organic solar cells could be a more practical selection because of its perfect compatibility with solution-processing, intrinsic flexibility and chemical stability. Especially with the introduction of non-fullerene acceptors, the efficiency of organic solar cells can reach beyond 18%, which is satisfying and promising.

[0005] Typically, depending on the type of organic donor-acceptor pair, the bandgaps of the perovskite layer in a perovskite/organic tandem solar cell may vary from around 1.78-1.92 eV. Perovskites at this bandgap range usually contain more than 30% by weight of bromide component, even up to 50% by weight of bromide component based on the total weight of the wide-bandgap perovskite layer, which will naturally induce large non-radiative losses and harmful halide phase segregation problem as compared to its narrow-bandgap perovskite counterparts. This creates an obstacle for further improvement of power conversion efficiency and long-term stability of perovskite-based solar cells, including perovskite/organic tandem solar cells.

[0006] It is therefore desirable to provide a solar cell having a perovskite-type active layer which seeks to address at least one of the problems described hereinabove, or at least to provide an alternative.

SUMMARY

[0007] According to a first aspect of the present disclosure, the present disclosure provides a solar cell for converting energy of light into electricity. The solar cell comprises a first subcell comprising: a transparent substrate; a first interlayer deposited on the transparent substrate; a second interlayer; and a wide-bandgap metal halide perovskite-type active layer having a bandgap greater than 1.8eV deposited between the first interlayer and the second interlayer for absorbing high-energy photons, wherein the wide-bandgap metal halide perovskite-type active layer includes 2-amino-4,5-imidazoledicarbonitrile (AIDCN) as an organic additive, represented by the following chemical formula (I):

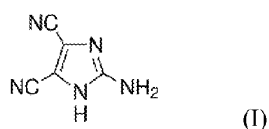


[0008] In some embodiments, the solar cell further comprises a second subcell comprising: a transparent conductive oxide layer; a third interlayer deposited on the second transparent substrate; an organic donor-acceptor complex material as an active layer deposited on the third interlayer for absorbing low-energy photons remaining after the energy of light passes through the first subcell; and a fourth interlayer deposited on the organic donor-acceptor complex material; wherein the first subcell and the second subcell are integrated to form a perovskite/organic tandem solar cell structure.

[0009] In some embodiments, the first subcell further comprises an electrode deposited on the second interlayer to form a single-junction wide-bandgap perovskite solar cell with a bandgap greater than 1.8eV.

[0010] According to a second aspect of the present disclosure, a perovskite/organic tandem solar cell is provided. The perovskite/organic tandem solar cell comprises a first subcell comprising a wide-bandgap metal halide perovskite-type active layer having a bandgap greater than 1.8eV for absorbing high-energy photons; a second subcell comprising an organic donor-

acceptor complex material as an active layer for absorbing low-energy photons remaining after the energy of light passes through the first subcell; and wherein the wide-bandgap metal halide perovskite-type active layer includes 2-amino-4,5-imidazoledicarbonitrile (AIDCN) as an organic additive represented by the following chemical formula (I):



[0011] In some embodiments, the first subcell further comprises a transparent substrate; a first hole transport layer deposited between the transparent substrate and the wide-bandgap metal halide perovskite-type active layer; and a first electron transport layer deposited on the wide-bandgap metal halide perovskite-type active layer.

[0012] In some embodiments, the second subcell further comprises a transparent conductive oxide layer deposited on the first electron transport layer; a second hole transport layer deposited on the transparent conductive oxide layer; and a second electron transport layer deposited on the organic donor-acceptor complex material, wherein the organic donor-acceptor complex is deposited on the second hole transport layer.

[0013] In some embodiments, the perovskite/organic tandem solar cell further comprises an electrode deposited on the second electron transport layer.

[0014] According to a third aspect of the present disclosure, a method for preparing a perovskite/organic tandem solar cell is provided. The method comprises cleaning a transparent substrate and carrying out a UV treatment for a duration of time; depositing a first hole transport layer on the transparent substrate; depositing a wide-bandgap metal halide perovskite-type active layer having a bandgap greater than 1.8eV, including 2-amino-4,5-imidazoledicarbonitrile (AIDCN) on the first hole transport layer; depositing an interconnecting layer on the wide-bandgap metal halide perovskite-type active layer; depositing an organic

donor-acceptor complex material as an active layer on the interconnecting layer; and depositing a second electron transport layer on the organic donor-acceptor complex material.

[0015] In some embodiments, the step of depositing the interconnecting layer on the wide-bandgap metal halide perovskite-type active layer comprises depositing a first electron transport layer on the wide-bandgap metal halide perovskite-type active layer; sputtering a transport conductive oxide layer on the first electron layer; and thermally evaporating a second hole transport layer on the transport conductive oxide layer.

BRIEF DESCRIPTION OF THE DRAWINGS

[0016] Various embodiments of the present disclosure are described hereinbelow in the detailed description with reference to the following drawings:

FIG. 1A is a schematic diagram of device configuration of a solar cell employing a single-junction wide-bandgap perovskite solar cell with a bandgap greater than 1.8 eV in accordance with various embodiments of the present disclosure.

FIG. 1B is a cross-sectional SEM image of an exemplary embodiment of the single-junction wide-bandgap perovskite solar cell with a bandgap greater than 1.8 eV of the present disclosure.

FIG. 2A is a schematic diagram of device configuration of a solar cell employing a perovskite/organic tandem solar cell in accordance with various embodiments of the present disclosure.

FIG. 2B is a cross-sectional SEM image of an exemplary embodiment of the perovskite/organic tandem solar cell of the present disclosure.

FIG. 3A is a plot showing the champion $J-V$ curves (in reverse scans) of the wide-bandgap perovskite solar cell.

FIG. 3B is a plot showing the champion $J-V$ curves in forward and reverse scan directions for the wide-bandgap perovskite solar cell.

FIG. 3C is a plot showing the EQE curves and integrated J_{sc} for the wide-bandgap perovskite solar cell.

FIG. 3D is a plot showing the photovoltaic bandgaps determined by inflection points of EQE curves.

FIG. 3E show the UV-Vis absorption spectra (i) and tauc-plots (ii) with fitted optical bandgaps for the wide-bandgap perovskite solar cell without and with AIDCN.

FIG. 4A shows the statistical distributions of PCE of the wide-bandgap perovskite solar cell.

FIG. 4B shows the statistical distributions of V_{oc} of the wide-bandgap perovskite solar cell.

FIG. 4C shows the statistical distributions of FF parameters of the wide-bandgap perovskite solar cell.

FIG. 5A, FIG. 5B show the statistical distributions of J_{sc} (A) and $V_{oc} \times FF$ products (B) of the wide-bandgap perovskite solar cell.

FIG. 6A and FIG. 6B are plots showing the comparisons between the wide-bandgap perovskite solar cell with a bandgap greater than 1.8 eV in accordance with various embodiments of the present disclosure and previous reports of 1.80-1.89 eV perovskite solar cell in terms of PCE vs bandgap (A) and FF vs V_{oc} (B).

FIG. 7 is a plot showing the HREQE curves of the wide-bandgap perovskite solar cell of the present disclosure.

FIG. 8A and FIG. 8B are plots showing the fitting Urbach energy for wide-bandgap perovskite solar cell (FIG. 8A: control perovskite, FIG. 8B: perovskite with AIDCN).

FIG. 9A and FIG. 9B are plots showing the fitting QFLS for wide-bandgap perovskite solar cell (FIG. 9A: control perovskite, FIG. 9B: perovskite with AIDCN).

FIG. 10 is a plot showing the V_{oc} loss analysis for perovskite without and with AIDCN.

FIG. 11 is a plot showing long-term MPP stability assessments following ISOS-L-1 for wide-bandgap perovskite solar cell with a bandgap greater than 1.8 eV.

FIG. 12 is a plot showing the EQE curves and integrated J_{sc} for perovskite and organic (OPV) subcell in perovskite/organic tandem solar cell, respectively.

FIG. 13 is a plot showing combination of champion $J-V$ curves and photovoltaic parameters of single-junction organic solar cell (P1) (OPV: V_{oc} : 0.852V, J_{sc} : 26.15 mA cm⁻², FF: 74.89% and PCE: 16.68%); single-junction perovskite solar cell (P2) (PVK: V_{oc} : 1.366 V, J_{sc} : 16.10 mA cm⁻², FF: 84.20% and PCE: 18.52%); and perovskite/organic tandem solar cell with AIDCN (P3) (PVK/OPV tandem solar cell: V_{oc} : 2.140 V, J_{sc} : 15.04 mA cm⁻², FF: 76.77% and PCE: 24.70%).

FIG. 14 is a plot showing the statistical distributions of PCE of the perovskite/organic tandem solar cell with AIDCN.

FIG. 15 is plot showing the certified $J-V$ curve and photovoltaic parameters of the perovskite/organic tandem solar cell with AIDCN (V_{oc} : 2.136 V, J_{sc} : 14.59 mA cm⁻², FF: 75.7%, PCE: 23.6% and Area: 0.0552 cm²).

FIG. 16A and FIG. 16B show the certification summarizing the performance of the perovskite/organic tandem solar cell with AIDCN.

FIG. 17 shows the certified MPP stability results and photovoltaic parameters after 300s MPP stability test of the perovskite/organic tandem solar cell with AIDCN (V_{max} : 1.735 V, J_{max} : 13.50 mA cm⁻², P_{max} : 1.293 mW, PCE: 23.4%, Area: 0.0552 cm² and temperature: 25±1°C).

FIG. 18A is a plot showing the I 3d XPS results of perovskite without and with AIDCN.

FIG. 18B and FIG. 18C are plots showing the N 1s XPS results of perovskite without (B) and with (C) AIDCN.

FIG. 18D is a plot showing the FTIR results of AIDCN, perovskite (PVK) without and with AIDCN.

FIG. 19A is a plot showing the calculated PLQY comparison for perovskite without and with AIDCN. Inset: steady-state PL spectra of perovskite without and with AIDCN.

FIG. 19B and FIG. 19C are contour plots showing PL evolutions of perovskite without (B) and with (B) AIDCN under continuous 532 nm laser excitation.

FIGS. 20A-20D show the hyperspectral imaging of normalized PL intensity (FIGS. 20A and 20C) and PL peak positions (FIGS. 20B and 20D) for the samples before aging (FIG. 20A, FIG. 20B: control, FIG. 20C, FIG. 20D: AIDCN). Scale bars: 50 μm .

FIGS. 21A and 21B show the hyperspectral imaging of normalized PL intensity of aged perovskite without (A) and with (B) AIDCN after 10 mins' 532 nm laser excitation.

FIGS. 22A and FIG. 22B show the hyperspectral imaging of PL peak position of aged perovskite without (A) and with (B) AIDCN after 10 mins' 532 nm laser excitation.

FIG. 23A is a sketch illustrating the mechanism of phase segregation happening in control perovskite (left) and suppressed phase segregation in perovskite with AIDCN (right).

FIG. 23B is a plot showing the UV-Vis absorptions of toluene solution with perovskite thin-films immersed after 24 hours light illumination.

FIGS. 24A-24D show the 2D GIWAXS maps of both conditions after 0 and 5 minutes of light soaking, respectively.

FIGS. 25A and 25B are plots showing the pseudo-XRD results of (200) diffraction of perovskite without (A) and with (B) AIDCN versus light soaking time.

FIG. 26 is a plot showing the evolution of fitted (200) peak position for perovskite from *in situ* GIWAXS measurements under continuous light soaking. "RP" refers to residual phase.

FIG. 27 is a plot showing the evolution of local I:Pb ratios extracted from *in situ* XRF mappings under continuous light soaking.

FIG. 28 is a plot showing the changes of local Br:Pb ratios extracted from *in situ* XRF mappings before and after 10 minutes' continuous light soaking.

DESCRIPTION

[0017] The following description sets forth exemplary methods, parameters, and the like. The embodiments are described in sufficient detail to enable those skilled in the art to practise the invention. Other embodiments may be utilized, and structural and logical changes may be made without departing from the scope of the invention. The various embodiments are not necessarily mutually exclusive, as some embodiments can be combined with one or more other embodiments to form new embodiments.

[0018] Features that are described in the context of an embodiment may correspondingly be applicable to the same or similar features in the other embodiments. Features that are described in the context of an embodiment may correspondingly be applicable to the other embodiments, even if not explicitly described in these other embodiments. Furthermore, additions and/or combinations and/or alternatives as described for a feature in the context of an embodiment may correspondingly be applicable to the same or similar feature in the other embodiments.

[0019] In the context of various embodiments, the articles “a”, “an” and “the” as used with regard to a feature or element include a reference to one or more of the features or elements.

[0020] In the context of various embodiments, the term “about” or “approximately” as applied to a numeric value encompasses the exact value and a reasonable variance, e.g. within 10% of the specified value.

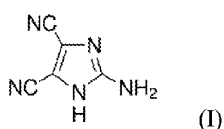
[0021] As used herein, the term “and/or” includes any, and all combinations of one or more of the associated listed items.

[0022] By “comprising” it is meant including, but not limited to, whatever follows the word “comprising”. Thus, use of the term “comprising” indicates that the listed elements are required or mandatory, but that other elements are optional and may or may not be present.

[0023] By “consisting of” is meant including, and limited to, whatever follows the phrase “consisting of”. Thus, the phrase “consisting of” indicates that the listed elements are required or mandatory, and that no other elements may be present.

[0024] Targeting on improving the power conversion efficiency (PCE), stability of solar cells and the halide phase segregation issue, a solar cell and a perovskite/organic tandem solar cell comprising an imidazole-based organic additive, in particular, 2-amino-4,5-imidazoledicarbonitrile (AIDCN) are developed. The imidazole-based organic additive is incorporated into a perovskite-type active layer of the solar cell and the perovskite/organic tandem solar cell to comprehensively enhance the photovoltaic performance of the solar cell and the perovskite/organic tandem solar cell.

[0025] In one aspect, a solar cell for converting energy of light into electricity is provided. With reference to FIG. 1A, the solar cell comprises a first subcell 100 comprising a transparent substrate 102; a first interlayer 104 deposited on the transparent substrate 102; a second interlayer 106; and a wide-bandgap metal halide perovskite-type active layer 108 having a bandgap greater than 1.8eV deposited between the first interlayer and the second interlayer for absorbing high-energy photons, wherein the wide-bandgap metal halide perovskite-type active layer includes 2-amino-4,5-imidazoledicarbonitrile (AIDCN) as an organic additive, represented by the following chemical formula (I):



[0026] In some embodiments, the first subcell further comprises an electrode 110 deposited on the second interlayer 106 to form a single-junction wide-bandgap perovskite solar cell with a bandgap greater than 1.8 eV. The electrode 110 may include a metal or a conductive material. Some examples include, but are not limited to, silver (Ag), gold (Au), indium zinc oxide (IZO)

and aluminium-doped zinc oxide (AZO). In some embodiments, the electrode is formed of silver. The electrode has a thickness ranging from 100 to 200 nm.

[0027] Unless otherwise defined, “wide-bandgap metal halide perovskite-type active layer” when used herein refers to a metal halide perovskite-type active layer having a bandgap greater than 1.8 eV.

[0028] Unless otherwise defined, “wide-bandgap perovskite” when used herein refers to a wide-bandgap perovskite having a bandgap greater than 1.8 eV.”

[0029] In some embodiments, the wide-bandgap metal halide perovskite-type active layer 108 is formed of a perovskite material comprising cesium (Cs), faradium (FA), lead (Pb), iodide (I) and bromine (Br) and 2-amino-4,5-imidazoledicarbonitrile (AIDCN) as the organic additive. The organic additive is incorporated into the perovskite-type active layer by adding and mixing the organic additive with a perovskite precursor solution containing formamidinium iodide (FAI), cesium iodide (CsI), lead (II) bromide (PbBr₂) and lead iodide (PbI₂). In some embodiments, the wide-bandgap metal halide perovskite-type active layer has a thickness ranging from 250 to 350 nm, preferably 280 to 320 nm, and preferably around 300 nm.

[0030] In some embodiments, the 2-amino-4,5-imidazoledicarbonitrile (AIDCN) incorporated into the wide-bandgap metal halide perovskite-type active layer has a concentration ranging from 1.0 to 2.0 mg/mL, preferably 1.3 to 1.7 mg/mL and more preferably 1.5mg/mL.

[0031] In some embodiments, the wide-bandgap metal halide perovskite-type active layer contains 40% to 50% bromide component based on the total weight of the wide-bandgap metal halide perovskite-type active layer. In some embodiments, the wide-bandgap metal halide perovskite-type active layer has a bandgap of 1.80 - 2.00 eV.

[0032] In some embodiments, the first interlayer 104 is a hole transport layer. The hole transport layer may be provided in the form of a single layer or two or more layers containing

highly hole-transportable substances. In some embodiments, the hole transport layer comprises nickel oxide and (4-(3,6-dimethyl-9H-carbazol-9-yl)butyl)phosphonic acid (Me-4PACz). This includes embodiments comprising a single layer made of nickel oxide and (4-(3,6-dimethyl-9H-carbazol-9-yl)butyl)phosphonic acid (Me-4PACz) and embodiments comprising two or more layers consisting of a layer made of nickel oxide and a layer made of (4-(3,6-dimethyl-9H-carbazol-9-yl)butyl)phosphonic acid (Me-4PACz).

[0033] In some embodiments, the second interlayer 106 is an electron transport layer. The electron transport layer may be provided in the form of a single layer or two or more layers comprising materials that have high electron mobility and high electron affinity. In some embodiments, the electron transport layer comprises (6,6)-phenyl-C₆₁-butyric acid methyl ester (PCBM) and 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP). This includes embodiments comprising a single layer made of (6,6)-phenyl-C₆₁-butyric acid methyl ester (PCBM) and 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) and embodiments comprising two or more layers consisting of a layer made of (6,6)-phenyl-C₆₁-butyric acid methyl ester (PCBM) and a layer made of 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP).

[0034] In some embodiments, the transparent substrate is a glass substrate or a patterned indium tin oxide (ITO) glass substrate.

[0035] Referring to FIG. 2A, the solar cell may further comprise a second subcell 200 comprising a transparent conductive oxide layer 202; a third interlayer 204 deposited on the transparent conductive oxide layer 202; an organic donor-acceptor complex material as an active layer 206 deposited on the third interlayer for absorbing low-energy photons remaining after the energy of light passes through the first subcell; and a fourth interlayer 208 deposited on the organic donor-acceptor complex material 206, wherein the first subcell 100 and the second subcell 200 are integrated to form a perovskite/organic tandem solar cell structure 201.

[0036] In some embodiments, the second subcell further comprises an electrode 210 deposited on the fourth interlayer 208. Any suitable electrode can be used without departing from the scope of the present invention. The electrode 210 may include a metal or a conductive material. Some examples include, but are not limited to, silver (Ag), gold (Au), indium zinc oxide (IZO) and aluminium-doped zinc oxide (AZO). In some embodiments, the electrode is formed of silver. The electrode has a thickness ranging from 100 to 200 nm.

[0037] Any suitable transparent conductive oxide layer 202 with high electron mobility may be used without departing from the scope of the present invention. Some examples include, but are not limited to, indium tin oxide (ITO), indium zinc oxide (IZO) and aluminium-doped zinc oxide (AZO). In some embodiments, the transparent conductive oxide layer is an indium tin oxide (ITO) layer.

[0038] In the embodiments comprising the second subcell, the second interlayer 106 is an electron transport layer and may comprise (6,6)-phenyl-C₆₁-butyric acid methyl ester (PCBM) and an aluminum-doped zinc oxide nanoparticle ink (AZO). This includes embodiments comprising a single layer made of (6,6)-phenyl-C₆₁-butyric acid methyl ester (PCBM) and an aluminum-doped zinc oxide nanoparticle ink (AZO), and embodiments comprising two or more layers consisting of a layer made of (6,6)-phenyl-C₆₁-butyric acid methyl ester (PCBM) and a layer made of an aluminum-doped zinc oxide nanoparticle ink (AZO).

[0039] In some embodiments, the third interlayer 204 is a second hole transport layer. The second hole transport layer may be provided in the form of a single layer or two or more layers containing highly hole-transportable substances. In some embodiments, the second hole transport layer is provided in the form of a single layer comprising molybdenum oxide (MoO_x).

[0040] In the embodiments comprising the second subcell, the second interlayer 106 may comprise (6,6)-phenyl-C₆₁-butyric acid methyl ester (PCBM). In this embodiment, the third interlayer 204 may form a single layer with the transparent conductive oxide layer 202 and the

layer is referred to herein as a “combined layer”. In one embodiment, the combined layer may comprise MoO_x, indium tin oxide (ITO) and aluminum-doped zinc oxide nanoparticle ink (AZO).

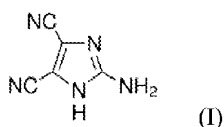
[0041] In some embodiments, the fourth interlayer 208 is a second electron transport layer. The second electron transport layer may be provided in the form of a single layer or two or more layers comprising materials that have high electron mobility and high electron affinity. In some embodiments, the second electron transport layer is made of N,N'-Bis{3-[3-(dimethylamino)propylamino]propyl}perylene-3,4,9,10-tetracarboxylic diimide (PDINN).

[0042] Any suitable organic donor-acceptor complex material may be used without departing from the scope of the present invention. In some embodiments, the organic donor-acceptor complex material is fabricated from poly[(2,6-(4,8-bis(5-(2-ethylhexyl-3-fluoro)thiophen-2-yl)-benzo[1,2-b:4,5-b']dithiophene))-alt-(5,5-(1',3'-di-2-thenyl-5',7'-bis(2-ethylhexyl)benzo [1',2-c:4',5'-c'dithiophene-4,5-dione]))] (PM6) and BTP-eC11 donor-acceptor system. The organic donor-acceptor complex material has a thickness ranging from 100 to 150 nm.

[0043] In some embodiments, the solar cell comprising the first subcell and the second subcell is configured in a monolithic two-terminal arrangement. Each of the first subcell and the second subcell is configured in a p-i-n arrangement.

[0044] In a second aspect, a perovskite/organic tandem solar cell is provided. The perovskite/organic tandem solar cell comprises a first subcell comprising a wide-bandgap metal halide perovskite-type active layer having a bandgap greater than 1.8eV for absorbing high-energy photons; a second subcell comprising an organic donor-acceptor complex material as an active layer for absorbing low-energy photons remaining after the energy of light passes through the first subcell; and wherein the wide-bandgap metal halide perovskite-type active layer

includes 2-amino-4,5-imidazoledicarbonitrile (AIDCN) as an organic additive represented by the following chemical formula (I):



[0045] In some embodiments, the first subcell further comprises a transparent substrate; a first hole transport layer deposited between the transparent substrate and the wide-bandgap metal halide perovskite-type active layer; and a first electron transport layer deposited on the wide-bandgap metal halide perovskite-type active layer.

[0046] In some embodiments, the second subcell further comprises a transparent conductive oxide layer deposited on the first electron transport layer; a second hole transport layer deposited on the transparent conductive oxide layer; and a second electron transport layer deposited on the organic donor-acceptor complex material, wherein the organic donor-acceptor complex is deposited on the second hole transport layer.

[0047] The perovskite/organic tandem solar cell further comprises an electrode deposited on the second electron transport layer.

[0048] In some embodiments, the transparent substrate comprises indium tin oxide (ITO); the first hole transport layer comprises nickel oxide and (4-(3,6-dimethyl-9H-carbazol-9-yl)butyl)phosphonic acid (Me-4PACz); and the first electron transport layer comprises (6,6)-phenyl-C₆₁-butyric acid methyl ester (PCBM) and aluminum-doped zinc oxide nanoparticle ink (AZO).

[0049] In some embodiments the transparent conductive oxide layer is made of indium tin oxide (ITO); the second hole transport layer is made of molybdenum oxide (MoO₃); and the second electron transport layer is made of N,N'-Bis{3-[3-(dimethylamino)propylamino]propyl}perylene-3,4,9,10-tetracarboxylic diimide (PDINN).

[0050] The first hole transport layer may be provided in the form of a single layer or two or more layers comprising materials having highly hole-transportable substances. In some embodiments, the first hole transport layer comprises a single layer made of nickel oxide and (4-(3,6-dimethyl-9H-carbazol-9-yl)butyl)phosphonic acid (Me-4PACz). In some embodiments, the first hole transport layer comprises two or more layers consisting of a layer made of nickel oxide and a layer made of (4-(3,6-dimethyl-9H-carbazol-9-yl)butyl)phosphonic acid (Me-4PACz).

[0051] In some embodiments, the first electron transport layer is provided in the form of a single layer or two or more layers comprising materials that have high electron mobility and high electron affinity. In some embodiments, the first electron transport layer comprises a single layer made of (6,6)-phenyl-C₆₁-butyric acid methyl ester (PCBM) and aluminum-doped zinc oxide nanoparticle ink (AZO). In some embodiments, the first electron transport layer comprises two or more layers consisting of a layer made of (6,6)-phenyl-C₆₁-butyric acid methyl ester (PCBM) and a layer made of aluminum-doped zinc oxide nanoparticle ink (AZO).

[0052] In a third aspect, a method for preparing a perovskite/organic tandem solar cell is provided. The method comprises cleaning a transparent substrate and carrying out a UV treatment for a duration of time; depositing a first hole transport layer on the transparent substrate; depositing a wide-bandgap metal halide perovskite-type active layer having a bandgap greater than 1.8 eV, including a 2-amino-4,5-imidazoledicarbonitrile (AIDCN) on the first hole transport layer; depositing an interconnecting layer on the wide-bandgap metal halide perovskite-type active layer; depositing an organic donor-acceptor complex material as an active layer on the interconnecting layer; and depositing a second electron transport layer on the organic donor-acceptor complex material.

[0053] In some embodiment, the wide-bandgap metal halide perovskite-type active layer including the 2-amino-4,5-imidazoledicarbonitrile (AIDCN) is prepared by adding the 2-

amino-4,5-imidazoledicarbonitrile (AIDCN) with a concentration of 1.5mg/mL into a perovskite precursor solution comprising 40% to 50% of bromide component; and performing spin coating to obtain the wide-bandgap metal halide perovskite-type active layer with the 2-amino-4,5-imidazoledicarbonitrile (AIDCN).

[0054] In some embodiments, the step of depositing the interconnecting layer on the wide-bandgap metal halide perovskite-type active layer comprises depositing a first electron transport layer on the wide-bandgap metal halide perovskite-type active layer; sputtering a transport conductive oxide layer on the first electron layer; and thermally evaporating a second hole transport layer on the transport conductive oxide layer. The method may further comprise depositing silver particles as an electrode on the second electron transport layer.

[0055] In various embodiments, the various layers of the solar cell and the perovskite/organic tandem solar cell are deposited by spin-coating or sputtering. One skilled in the art will appreciate that other suitable deposition methods may be used without departing from the scope of the present disclosure.

[0056] In the present disclosure, the addition of 2-amino-4,5-imidazoledicarbonitrile (AIDCN) to the wide-bandgap metal halide perovskite-type active layer helps to increase the power conversion efficiency (PCE) of the single-junction wide-bandgap perovskite solar cell to about 18.52% for a 1.86eV wide-bandgap perovskite solar cell with T_{80} stability of 271 hours at maximum power point and the PCE of the perovskite/organic tandem solar cell to about 24.70% (23.60% certified).

[0057] The 2-amino-4,5-imidazoledicarbonitrile (AIDCN) molecule has unprecedentedly high dipole moment with functional groups that could interact with the perovskite components in the perovskite-type active layer. By incorporating the AIDCN into the perovskite-type active layer, the solar cell reduces its non-radiative losses by passivating bulk defects (by various interactions between the AIDCN and the active layer components), reducing mobile chemical

species and thus leading to suppressed phase segregation. The AIDCN molecule with highly polarized distribution of charges can effectively interact with the iodide in the perovskite-type active layer, leading to mitigated photoinduced iodide oxidation, thus helps to suppress the phase segregation fundamentally.

[0058] The perovskite/organic tandem solar cell with the AIDCN can provide higher power conversion efficiency (PCE) which could help generate more electricity under same illumination spectrum. It also has better stability, thus leading to extended lifetime of the perovskite/organic tandem solar cell, which effectively reduces the costs of fabrication and installation of new devices comprising the perovskite/organic tandem solar cell. The perovskite/organic tandem solar cell is effective and compatible for application across all suitable technical fields which take advantage of perovskite/organic tandem solar cell as energy conversion device.

[0059] To facilitate a better understanding of the present disclosure, the following examples of specific embodiments are given. In no way should the following examples be read to limit or define the entire scope of the disclosure. One skilled in the art will recognize that the examples set out below are not an exhaustive list of the embodiments of this disclosure.

EXAMPLES

Materials

[0060] 2-amino-4,5-imidazoledicarbonitrile (AIDCN, 97%), cesium iodide (CsI, 99.999%), *N,N*-dimethylformamide (DMF, anhydrous), dimethyl sulfoxide (DMSO, anhydrous), methyl acetate (MeAc, anhydrous), 2-propanol (IPA, anhydrous), chlorobenzene (CB, anhydrous), dimethyl sulfoxide-*d*₆ (DMSO-*d*₆) and aluminum-doped zinc oxide nanoparticle ink (AZO, 2.5 wt.% in 1-butanol) were purchased from Sigma-Aldrich. Formamidineum iodide (FAI) was purchased from Greatcell Solar Materials. Lead iodide (PbI₂, 99.99%), lead bromide (PbBr₂) and [4-(3,6-Dimethyl-9H-carbazol-9-yl)butyl]phosphonic acid (Me-4PACz) were purchased

from Tokyo Chemical Industry. Silver pellets were purchased from Alfa Aesar and (6,6)-phenyl-C61-butyric acid methyl ester (PCBM) and 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) were purchased from Luminescence Technology.

Device characterizations

[0061] The completed devices were masked with metal aperture masks (0.055 cm²) for J - V measurement by a Keithley 2400 source meter under simulated 1-sun AM 1.5G illumination (100 mW cm⁻²) with an ABET Technologies Sun 2000 solar simulator. The J - V measurements were performed in a N₂-filled glovebox. MPP stability tests of the devices were performed with MPP Tracking - 4B source measure unit system (Shenzhen Lancheng Technology Ltd.) with 100 mW cm⁻² white LED. EQE measurements were conducted using a Bentham PVE300-IVT system. The intensity of LED laser used was calibrated with built-in silicon and germanium diodes before measurements. For the HREQE tests, the light was chopped at 137 Hz and coupled into a Bentham monochromator. The monochromatic light spot was focused onto the active area of perovskite solar cell, and its current under short-circuit conditions was fed to a current preamplifier (Stanford SR 570) before it was analyzed with a lock-in amplifier (Stanford SR830 DSP). The time constant of the lock-in amplifier was chosen to be 1s and the amplification of the preamplifier was increased to resolve low photocurrents. The EQE was determined by dividing the photocurrent of the cell by the flux of incoming photons, which was measured using a calibrated Si photodiode. All EQE measurements were conducted at ambient atmosphere and room temperature with a relative humidity of around 60% with encapsulation. Cross-section SEM images of devices were taken with Regulus SU8200 system (Hitachi) at 5 kV accelerating voltage under SE mode.

Material Characterizations

[0062] UV-Vis absorbance spectra were recorded with Agilent Cary 7000 spectrophotometer. The lamp was pre-heated for 30 min before conducting measurements.

Steady-state photoluminescence (PL) and PL evolution measurements were done by LP20-32 radiative efficiency meter (Quantum Yield Berlin). The built-in 532 nm laser was switched on in advance to allow warmup and stabilization. All of PL spectra were recorded under 1-sun 532 nm laser illumination. XRD is recorded on an X-ray diffractometer (Malvern Panalytical) with Cu K α radiation ($\lambda = 0.1542$ nm).

Example 1 – Fabrication of single-junction wide-bandgap perovskite solar cell with a bandgap greater than 1.8eV (without 2-amino-4,5-imidazoledicarbonitrile (AIDCN))

[0063] A single-junction wide-bandgap perovskite solar cell based on an inverted p-i-n structure and with a bandgap greater than 1.8eV was fabricated as described hereinbelow.

[0064] Patterned indium tin oxide (ITO) glass substrates were cleaned with surfactant solution, then soaked in deionized water, acetone and IPA in the ultrasonic bath for 15 min each. After ultraviolet ozone treatment for 15 min, the substrates were then spin-coated with nickel oxide (NiO_x) (10 mg/ml in deionized water) nanoparticle dispersion. The NiO_x films were annealed at 120°C for 10 min. Then the films were transferred to a nitrogen-filled glovebox and Me-4PACz ([4-(3,6-dimethyl-9H-carbazol-9-yl)butyl]phosphonic acid) (1 mg/ml in IPA) was spin-coated on the substrates at 4,000 rpm for 30s and heated at 100°C for 10 min. A 1 M perovskite (Cs_{0.25}FA_{0.75}Pb(I_{0.5}Br_{0.5})₃) precursor solution containing 129mg FAI, 65mg CsI, 275mg PbBr₂ and 115mg PbI₂ was dissolved in 1 ml mixed solvent of DMF:DMSO (V/V = 4:1). The precursor solution was then shaken overnight at 60°C. For spin-coating process, the substrate was spun at 4,000 rpm for 45s at an acceleration of 4000 rpm/s and 200 μ L of MeAc was slowly dropped at 15s before the end of the spin-coating. The perovskite films were then annealed at 100°C for 15 minutes. Next, PCBM ((6,6)-phenyl-C₆₁-butyric acid methyl ester) (20 mg/mL in CB) was spin-coated onto the perovskite thin-films at 1,000 rpm for 30s with an acceleration of 500 rpm/s, then annealed at 100°C for 5 min. Finally, BCP solution (2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline) (saturated in IPA) was dynamically

spin-coated onto the PCBM films at 5,000 rpm and annealed at 90°C for 2 min. Then the films were transferred to the thermal evaporator where 100nm Ag was deposited to complete the device fabrication. This device configuration is as shown in FIG. 1A, which is indium tin oxide 102 (ITO) / nickel oxide 104 (NiO_x) / [4-(3,6-Dimethyl-9H-carbazol-9-yl)butyl]phosphonic acid 104 (Me-4PACz) / 1.86 eV Cs_{0.25}FA_{0.75}Pb(I_{0.5}Br_{0.5})₃ wide-bandgap perovskite 108 / (6,6)-phenyl-C₆₁-butyric acid methyl ester 106 (PCBM) / 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline 106 (BCP) / Ag 110. The cross-sectional scanning electron microscope (SEM) image of this device configuration is as shown in FIG. 1B.

Example 2 – Fabrication of single-junction wide-bandgap perovskite solar cell with a bandgap greater than 1.8eV and with 2-amino-4,5-imidazoledicarbonitrile (AIDCN)

[0065] A single-junction perovskite solar cell with a bandgap greater than 1.8eV and with AIDCN was fabricated in the same manner as described in Example 1 except that the precursor solution contains about 1.5 mg/mL AIDCN in addition to 129 mg FAI, 65 mg CsI, 275 mg PbBr₂ and 115 mg PbI₂, all dissolved in 1 ml mixed solvent of DMF:DMSO (V/V = 4:1).

[0066] To check the reproducibility of the device performances, several samples were prepared. FIG. 3A and FIG. 3B show the champion *J-V* characteristics and Table 1 below summarizes the samples' photovoltaic parameters.

Table 1 | Summary of photovoltaic parameters of champion devices.

Conditions	Scan Direction	V_{oc} / V	J_{sc} / mA cm^{-2}	FF / %	PCE / %
Control	Forward	1.329	15.69	82.79	17.26
	Reverse	1.323	15.58	80.86	16.67
AIDCN	Forward	1.361	15.92	85.24	18.47
	Reverse	1.366	16.10	84.20	18.52

[0067] Compared to the control, the power conversion efficiency (PCE) of the single-junction wide-bandgap perovskite solar cell with a bandgap greater than 1.8 eV and with AIDCN can be boosted from 17.26% to 18.52%, resulting from comprehensively enhanced V_{oc} , short-circuit current density (J_{sc}) and fill factor (FF). Results of external quantum efficiency (EQE) measurements pointed out a generally better photovoltaic response of the perovskite with AIDCN as compared to the control. This proves that the J_{sc} values obtained from the J - V scans were within 5% error with respect to the integrated J_{sc} from EQE (FIG. 3C). The photovoltaic bandgaps determined from the inflection points of the EQE curves (FIG. 3D) were in consistency with the optical bandgaps (FIG. 3E(i) and 3E(ii)) and it is concluded that the introduction of AIDCN will not alter the bandgap of perovskite. As a result, the V_{oc} improvement that was observed was from the mitigation of V_{oc} losses of the wide-bandgap perovskite solar cell (with a bandgap greater than 1.8 eV), which is another benefit brought by introducing AIDCN to the wide-bandgap perovskite solar cell with a bandgap greater than 1.8eV.

[0068] Several independent devices for each condition in the same batch for comparison were prepared and the results are as shown in FIG. 4A, FIG. 4B and FIG. 3C. The statistical distributions of these devices show better PCE, from around 17.26% to 18.52%, better V_{oc} from around 1.33V to 1.37V, better J_{sc} and a slightly improved distributions of FF for the wide-bandgap perovskite solar cell (with a bandgap greater than 1.8eV) with AIDCN as compared to the control, yielding an increased $V_{oc} \times FF$ product (FIG. 5A, FIG. 5B). Compared to previous reports of 1.80 eV to 1.89 eV wide-bandgap perovskite solar cells, this performance is among one of the best compared to 70% of S-Q limit as summarized in FIG. 6A. An unprecedentedly outstanding FF of more than 85% as well as one of the highest V_{oc} of 1.366 V for single-junction wide-bandgap perovskite solar cell has been achieved (FIG. 6B) in this specific

bandgap range of greater than 1.8 eV, which is particularly suitable for fabricating perovskite/organic or perovskite/perovskite tandem solar cells.

Example 3

To better understand the origin of the high V_{OC} realized, a high-resolution external quantum efficiency (HREQE) characterization was conducted to extract the radiative-limit $V_{OC}(V_{OC,rad})$ and to derive the Urbach energy of the devices (FIG. 7, FIG. 8). The fitted Urbach energy for perovskite with AIDCN is 17.59 eV, which is slightly lower than the 18.34 eV of control, indicating reduced electronic disorders in the sub-bandgap region. Quasi-fermi level splitting (QFLS) was also obtained from analyzing steady-state PL results (FIG. 9). Combining QFLS with the extracted $qV_{OC,rad}$ and the measured champion qV_{OC} of the devices, origins of high V_{OC} of the wide-bandgap perovskite solar cell (with a bandgap greater than 1.8eV) of the present disclosure were uncovered (FIG. 10). It could be concluded that the 37 meV improvement of device V_{OC} by introducing AIDCN is mainly from the reduced losses between the $qV_{OC,rad}$ and QFLS, from 196 meV to 167 meV. This difference of QFLS for the control and the perovskite with AIDCN confirmed the ability of AIDCN to improve the quality of wide-bandgap perovskite at the same time of suppressing phase segregation, which leads to reduced V_{OC} losses for the wide-bandgap perovskite solar cell (with a bandgap greater than 1.8 eV), which is favorable for fabrication of highly efficient perovskite solar cells and tandem solar cells.

[0069] Furthermore, as shown in FIG. 11, the long-term stabilities of wide-bandgap perovskite solar cell with a bandgap greater than 1.8eV were tracked at maximum power point (MPP) following ISOS-L-1 standard using 100 mW cm⁻² white LED at ambient conditions with encapsulation. The control wide-bandgap perovskite solar cell with a bandgap greater than 1.8 eV could maintain 80% of the initial PCE for 154 hours (T_{80}). However, with AIDCN incorporated, the T_{80} of the device could be extended to 271 hours, which is a 76% of increment compared to the reference. This suggests the positive effect of suppressed phase segregation on

the ISOS-L-1 MPP stabilities of solar cell devices, which is desirable considering that stability would be one of the main concerns for commercialization of the perovskite-based solar cells.

Example 4 – Fabrication of perovskite/organic tandem solar cell with AIDCN

[0070] Perovskite/organic tandem solar cells were fabricated to convert the positive doping effect of AIDCN in wide-bandgap perovskite (with a bandgap greater than 1.8 eV) to improve performance of tandem solar cell.

[0071] After completing deposition of PCBM layer as described in Example 1 and using the the precursor solution that contains about 1.5 mg/mL AIDCN in addition to 129mg FAI, 65mg CsI, 275mg PbBr₂ and 115mg PbI₂ as described in Example 2, the thin-film was dynamically spin-coated at 4,000 rpm, with AZO nanoparticle ink. It was then annealed at 100°C for 10 min before transferring to a sputter system where about 5nm of ITO was sputtered as a charge recombination layer. After which, 30 nm MoO_x was thermally evaporated on top of the ITO layer and the films were transferred back to the nitrogen-filled glovebox. Subsequently, an organic (OPV) precursor solution (PM6: BTP-eC11 with D/A=1:1.2 at a concentration of 8 mg/mL in chloroform) was spin-coated onto the films immediately after the addition of 0.5% 1,8-diiodooctane (V/V) to the precursor, then the films were annealed at 100°C for 10 min. The final thickness of the organic (OPV) active layer is around 100 nm. After which, PDINN (N,N'-Bis{3-[3-(Dimethylamino)propylamino]propyl}perylene-3,4,9,10-tetracarboxylic diimide) solution (1 mg/mL in methanol) was spin-coated at 3,000 rpm for 30s. Lastly, the films were transferred to a thermal evaporator where 100nm of Ag was deposited onto the PDINN layer as an electrode to form a perovskite/organic tandem solar cell with AIDCN.

[0072] FIG. 2A illustrates the device configuration of the perovskite/organic tandem solar cell with AIDCN, which comprises ITO 102 / NiO_x 104 / Me4PACz 104 / wide-bandgap perovskite (with a bandgap greater than 1.8 eV and with 1.5 mg/mL AIDCN) 108 / PCBM 106 / aluminum-doped zinc oxide 106 (AZO) / ITO 202 / molybdenum oxide 204 (MoO_x) / organic

layer 206 (OPV) / 2,9-Bis[3-[[3-(dimethylamino)propyl]amino]propyl]-anthra[2,1,9-def:6,5,10-d'e'f']diisoquinoline-1,3,8,10(2H,9H)-tetrone 208 (PDINN) / Ag 210. The cross-sectional scanning electron microscope (SEM) image of this device configuration is as shown in FIG. 2B.

Example 5

From the result of the EQE measurements of the perovskite/organic tandem solar cell with the AIDCN, it was proven that the bandgap of greater than 1.8 eV of the wide-bandgap perovskite in the present disclosure fits perfectly with the organic (OPV) layer according to the current matching rule of 2-terminal tandem solar cell (FIG. 12). The champion PCE of the perovskite/organic tandem solar cell with the AIDCN reached 24.70%, with V_{OC} of 2.140 V, J_{SC} of 15.04 mA cm⁻² and FF of 76.77%. It is one of the highest, if not the highest PCE reported for perovskite/organic tandem solar cell so far, confirming that the enhanced performance of the wide-bandgap perovskite (with bandgap greater than 1.8 eV) with AIDCN could be successfully transferred to improve perovskite/organic tandem solar cell. Champion $J-V$ curves were plotted as well as photovoltaic parameters for single-junction organic, single-junction perovskite solar cell and perovskite/organic tandem solar cell, respectively, as shown in FIG. 13 for comparison. The statistical distributions of PCE from 25 independent perovskite/organic tandem solar cells with AIDCN devices were also provided and as shown in FIG. 14. It generally follows a normal distribution centered at about 24.0% of PCE. The perovskite/organic tandem solar cell with AIDCN of the present disclosure was sent to an independent third-party agency, Japan Electrical Safety and Environment Technology Laboratories (JET) for test and the perovskite/organic tandem solar cell with AIDCN was certified with a PCE of 23.6%, with a V_{OC} of 2.136 V, J_{SC} of 14.59 mA cm⁻² and FF of 75.7% (FIG. 15, FIG. 16A). The PCE obtained in the present disclosure is one of the highest, if not the highest among all of the reported certified PCE for perovskite/organic tandem solar cells. This shows the great benefit gained by

the introduction of AIDCN. Furthermore, as shown in FIG. 17 and FIG. 16B, MPP stability was also tested for the certified perovskite/organic tandem solar cell of the present disclosure and within 300s continuous tracking, the perovskite/organic tandem solar cell could still maintain a high PCE of 23.4%, indicating superior stability of the perovskite/organic tandem solar cell enabled by AIDCN incorporation.

Example 6

[0073] A test was carried out to show the interaction between AIDCN and perovskite. The interaction from the iodide side of the perovskite was investigated through X-ray photoelectron spectroscopy (XPS). A wide-bandgap perovskite comprising $\text{Cs}_{0.25}\text{FA}_{0.75}\text{Pb}(\text{I}_{0.5}\text{Br}_{0.5})_3$ with an optical bandgap of 1.86 eV was selected as the default composition for this test (FIG. 3E). In FIG. 18A, I 3d XPS peaks were presented, and it clearly shows the shift to lower binding energy induced by the addition of AIDCN to the perovskite as compared to perovskite itself. It can be understood that in the first case, some AIDCN replaced the more positive Pb^{2+} to bond with iodide, leading to an increased electron density of iodide, which makes it more reductive against the photoinduced oxidation. The successful incorporation of AIDCN to the wide-bandgap perovskite (with a bandgap greater than 1.8 eV) was also confirmed by XPS and Fourier-transform infrared spectroscopy (FTIR) (FIG. 18B, FIG. 18C and FIG. 18D).

Example 7 - Suppression effect of phase segregation for perovskite with AIDCN

[0074] A test was carried out to show how the interaction between the AIDCN and perovskite contributes to the suppression of iodide oxidation reaction in the solar cell.

[0075] Firstly, a steady-state photoluminescence (PL) was performed and as illustrated in the inset of FIG. 19A, the intensity of PL emission peak for perovskite with AIDCN increased by more than twice compared to the control, suggesting a reduced non-radiative recombination in the perovskite. The PL quantum yield (PLQY) for both conditions was calculated, and it led to a significant enhancement from 0.043% for the control to 0.134% for the perovskite with

AIDCN incorporated. Subsequently, the suppression effect of phase segregation by incorporating AIDCN was investigated. In FIG. 19B and FIG. 19C, the evolution of PL was tracked for both of the conditions by exposing perovskite thin-films to continuous 532 nm laser illumination. The control sample revealed an obvious phase segregation, evidenced by a new PL peak formed which was centered at about 730 nm after 2 minutes, and then it gradually saturated around 5 minutes and thereafter, corresponding to emissions from segregated iodide-rich phase. On the sharp contrary, the perovskite sample with AIDCN showed no signs of phase segregation within 10 minutes of aging, suggesting a complete elimination of phase segregation in terms of PL evolution. Furthermore, a hyperspectral imaging to gain more spatial information of the segregation process was also performed. As shown in FIGS. 20A to 20D, perovskite with AIDCN shows a more homogenous distribution of normalized PL intensity, and both samples have similar single-peak PL emission features. However, after 10 minutes of aging, as illustrated in FIGS. 21A, 21B and FIGS. 22A and FIG. 22B, there were domains with stronger emission but at longer wavelength generated, which is consistent with previous observations of phase segregation. For perovskite doped with AIDCN, there is no detectable sign of phase segregation, which is in line with the above findings in the PL evolution tests conducted. The above tests demonstrate that there is indeed an obvious suppression effect of phase segregation brought by the AIDCN to the wide-bandgap perovskite-type active layer (with a bandgap of greater than 1.8 eV) of the present disclosure.

[0076] Various characterizations were employed to reveal the mechanism of phase segregation and how AIDCN can help to suppress this phenomenon.

[0077] FIG. 23A depicts the proposed mechanism which further develops Kerner *et al.*'s (Kerner *et al.*, "The role of halide oxidation in perovskite halide phase separation", *Perspective*, vol. 5, issue 9, P2273-2295, 15 September 2021) previous model based on some observations. FIG. 23A is a sketch illustrating a phase segregation happening in control

perovskite without AIDCN (left). The figure includes an original phase 231 comprising perovskite without AIDCN, a segregated phase 232, a residual phase 233 and a lattice re-entrance 234. The figure further illustrates the processes of photoinduced iodide oxidation 235, iodine diffusion 236 and iodine migration 237 that could take place in the proposed mechanism.

[0078] FIG. 23A (right) illustrates a phase segregation in perovskite with AIDCN (right). The figure illustrates a suppressed photoinduced iodide oxidation 238 after photoinduced iodide oxidation took place.

[0079] At the beginning, the presence of photoinduced iodide oxidation 235 was confirmed by immersing perovskite thin-films in 5 mL toluene, and then exposed to 24 hours' continuous 1-sun white LED soaking (FIG. 23B). The UV-Vis results for the aged toluene solution show signs of iodine formation, which has an absorption peak around 500 nm. It turned out that AIDCN could help reduce the amount of accumulated iodine to a near negligible level, compared to the control. Then, a series of *in situ* experiments were prepared to uncover the dynamic changes of the crystal structure and local chemical compositions of the perovskite in response to continuous light soaking. As shown in FIGS. 24A-24D, 2D grazing-incidence wide-angle X-ray scattering (GIWAXS) maps were plotted for both conditions at moments of beginning of light illumination and after 5 minutes of soaking. By extracting the pseudo-XRD results with respect to the soaking time (FIGS. 25A and 25B) and fitting the position of (200) peak (FIG. 26), we visualized a dynamic change of "averaged" crystal structure of perovskite during light illumination. As illustrated, the perovskite with AIDCN shows relatively constant peak position at around 2.042 Å within 20 minutes of soaking while for the perovskite control, it experienced a dramatic shift to higher q direction in the first 5 minutes, reflecting a decreased "averaged" lattice parameter in this case. This unusual phenomenon is ascribed to the formation of a kind of residual phase, with slightly more bromide ratio caused by the evaporation of iodine formed during light illumination. In 8 minutes, the (200) peak returned to its original position,

which can be explained by a balance created between iodine formation and iodide migration 237 outside the illumination region back to the residual phases 233, as illustrated in FIG. 23A. Therefore, the photoinduced oxidation of iodide to form iodine in the early stage of phase segregation can be acknowledged as the trigger for the whole process. This can be further verified by *in situ* X-ray fluorescence (XRF) measurements employing synchrotron X-ray beams. Br, I and Pb (from L emission) elements were identified from the raw spectra of XRF of perovskite samples and the relative ratio of I:Pb (FIG. 27) and Br:Pb (FIG. 28) ratios in different soaking times from maps of relevant elements were plotted. FIG. 28 shows the difference of local Br:Pb ratios between before and after first 10 minutes of light soaking, in which both conditions show very similar changes, possibly due to high intensity X-ray beams. While for local I:Pb ratio illustrated in FIG. 27, a contrast is observed compared to Br:Pb in the first 10 minutes, confirming that the introduction of AIDCN majorly interacts with iodide, instead of bromide in perovskite. Furthermore, the sharpest difference between both conditions appears in the first 10 minutes of light soaking. This is consistent with the GIWAXS measurements. It can be concluded that the incorporation of AIDCN could help maintain I:Pb ratio as compared to the control and this is in line with the observation and inference that it suppressed the photoinduced oxidation of iodide, as shown in FIG. 23A. Hence, we can now correlate the findings in PL evolution, *in situ* GIWAXS and XRF measurements. At approximately the first 10 minutes' scale of light soaking, the wide-bandgap perovskite (with a bandgap greater than 1.8 eV) will suffer from a photoinduced oxidation process of iodide 235, creating iodine vapor and leaving residual phases 233 with slightly more bromide ratio. Then diffusion of iodine 236, its re-entrance to lattice 234 and migration 237 will offset the effect of iodine formation on these residual phases 233 and the phase segregation saturated after around 10 minutes of light soaking. The origin of why AIDCN can suppress the phase segregation is now elucidated, which comes from its ability to mitigate the photoinduced iodide oxidation

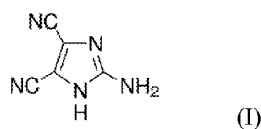
process, the first step of phase segregation, as illustrated in FIG. 23A.

[0080] The above examples demonstrate that the goal of addressing halide phase segregation issue in single-junction wide-bandgap perovskite solar cell with a bandgap greater than 1.8 eV and in wide-bandgap perovskite (with a bandgap greater than 1.8 eV) suitable for tandem solar cell can be fulfilled by adding an organic additive, namely AIDCN to the wide-bandgap metal halide perovskite-type active layer of the solar cell to induce chemical interactions with the iodide in the perovskite-type active layer of the solar cell. By various *in situ* characterizations, the examples demonstrated that AIDCN could help mitigate photoinduced iodide oxidation which is confirmed as the trigger of phase segregation. A single-junction wide-bandgap perovskite solar cell with a bandgap greater than 1.8 eV and with AIDCN is able to achieve a PCE of 18.52% with an improved V_{OC} to 1.366 V and a superior T_{80} of 271 hours following ISOS-L-1 standard of MPP tracking. The optimized perovskite in the wide-bandgap perovskite solar cell can be applied to perovskite/organic tandem solar cell and achieved a record high PCE of 24.70% (23.60% certified). These results demonstrate the effectiveness of the design strategy of the single-junction wide-bandgap perovskite solar cell (with a bandgap greater than 1.8 eV) and the perovskite/organic tandem solar cell of the present disclosure for additives to mitigate photoinduced iodide oxidation and enlighten more methods to suppress phase segregation from a fundamental perspective, contributing to improvements of efficiencies and stabilities for perovskite-based tandem solar cells.

[0081] Although embodiments of the invention have been shown and described, the invention is not limited to the described embodiments. Instead, it would be appreciated by those skilled in the art that various modifications and variations can be made to the embodiments of the invention without departing from the scope of the invention, the scope of which is set forth in the following claims.

Claims

1. A solar cell for converting energy of light into electricity, comprising:
- a first subcell comprising:
- a transparent substrate;
 - a first interlayer deposited on the transparent substrate;
 - a second interlayer; and
 - a wide-bandgap metal halide perovskite-type active layer having a bandgap greater than 1.8 eV, deposited between the first interlayer and the second interlayer for absorbing high-energy photons, wherein the wide-bandgap metal halide perovskite-type active layer includes 2-amino-4,5-imidazoledicarbonitrile (AIDCN) as an organic additive, represented by the following chemical formula (I):



2. The solar cell according to claim 1, further comprising:
- a second subcell comprising:
- a transparent conductive oxide layer;
 - a third interlayer deposited on the second transparent substrate;
 - an organic donor-acceptor complex material as an active layer deposited on the third interlayer for absorbing low-energy photons remaining after the energy of light passes through the first subcell; and
 - a fourth interlayer deposited on the organic donor-acceptor complex material;
- wherein the first subcell and the second subcell are integrated to form a perovskite/organic tandem solar cell structure.

3. The solar cell according to claim 1 or 2, wherein the first interlayer is a hole transport layer.
4. The solar cell according to claim 3, wherein the hole transport layer comprises nickel oxide and (4-(3,6-dimethyl-9H-carbazol-9-yl)butyl)phosphonic acid (Me-4PACz).
5. The solar cell according to claim 1, wherein the second interlayer is an electron transport layer.
6. The solar cell according to claim 5, wherein the electron transport layer comprises (6,6)-phenyl-C₆₁-butyric acid methyl ester (PCBM) and 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP).
7. The solar cell according to claim 2, wherein the second interlayer is an electron transport layer comprising (6,6)-phenyl-C₆₁-butyric acid methyl ester (PCBM) and an aluminum-doped zinc oxide nanoparticle ink (AZO).
8. The solar cell according to claim 2 or 7, wherein the third interlayer is a second hole transport layer.
9. The solar cell according to claim 8, wherein the second hole transport layer is made of molybdenum oxide (MoO_x).
10. The solar cell according to claim 2 or 9, wherein the fourth interlayer is a second electron transport layer.

11. The solar cell according to claim 10, wherein the second electron transport layer (ETL) is made of N,N'-Bis{3-[3-(dimethylamino)propylamino]propyl}perylene-3,4,9,10-tetracarboxylic diimide (PDINN).
12. The solar cell according to claim 1 or 2, wherein the transparent substrate and the transparent conductive oxide layer is made of indium tin oxide (ITO).
13. The solar cell according to claim 1, wherein the first subcell further comprises an electrode deposited on the second interlayer to form a single-junction wide-bandgap perovskite solar cell with a bandgap greater than 1.8 eV.
14. The solar cell according to claim 2 or 11, wherein the second subcell further comprises an electrode deposited on the fourth interlayer.
15. The solar cell according to any one of the preceding claims, wherein the wide-bandgap metal halide perovskite-type active layer has a bandgap of 1.80 - 2.00 eV.
16. The solar cell according to any one of the preceding claims, wherein the 2-amino-4,5-imidazoledicarbonitrile (AIDCN) has a concentration of 1.5mg/mL.
17. The solar cell according to any one of the preceding claims, wherein the wide-bandgap metal halide perovskite-type active layer contains 40% to 50% of bromide component based on the total weight of the wide-bandgap metal halide perovskite-type active layer.

18. The solar cell according to claim 2, wherein the solar cell is configured in a monolithic two-terminal arrangement.

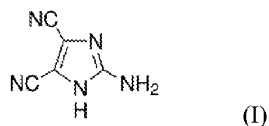
19. The solar cell according to claim 2, wherein each of the first subcell and the second subcell is configured in a p-i-n arrangement.

20. A perovskite/organic tandem solar cell comprising:

a first subcell comprising a wide-bandgap metal halide perovskite-type active layer having a bandgap greater than 1.8 eV for absorbing high-energy photons;

a second subcell comprising an organic donor-acceptor complex material as an active layer for absorbing low-energy photons remaining after the energy of light passes through the first subcell; and

wherein the wide-bandgap metal halide perovskite-type active layer includes 2-amino-4,5-imidazoledicarbonitrile (AIDCN) as an organic additive represented by the following chemical formula (I):



21. The perovskite/organic tandem solar cell according to claim 20, wherein the first subcell further comprises:

a transparent substrate;

a first hole transport layer deposited between the transparent substrate and the wide-bandgap metal halide perovskite-type active layer; and

a first electron transport layer deposited on the wide-bandgap metal halide perovskite-type active layer.

22. The perovskite/organic tandem solar cell according to claim 20 or 21, wherein the second subcell further comprises:
- a transparent conductive oxide layer deposited on the first electron transport layer;
 - a second hole transport layer deposited on the transparent conductive oxide layer; and
 - a second electron transport layer deposited on the organic donor-acceptor complex material, wherein the organic donor-acceptor complex is deposited on the second hole transport layer.
23. The perovskite/organic tandem solar cell according to claim 22, wherein the perovskite/organic tandem solar cell further comprises an electrode deposited on the second electron transport layer.
24. The perovskite/organic tandem solar cell according to any one of the preceding claims claim 20 to 23, wherein the 2-amino-4,5-imidazoledicarbonitrile (AIDCN) has a concentration of 1.5mg/mL.
25. The perovskite/organic tandem solar cell according to claim 21, wherein
- the transparent substrate is made of indium tin oxide (ITO);
 - the first hole transport layer comprises nickel oxide and (4-(3,6-dimethyl-9H-carbazol-9-yl)butyl)phosphonic acid (Me-4PACz); and
 - the first electron transport layer comprises (6,6)-phenyl-C₆₁-butyric acid methyl ester (PCBM) and aluminum-doped zinc oxide nanoparticle ink (AZO).
26. The perovskite/organic tandem solar cell according to claim 22, wherein the

the transparent conductive oxide layer is an indium tin oxide (ITO) layer;
the second hole transport layer is made of molybdenum oxide (MoO_x); and
the second electron transport layer is made of N,N'-Bis{3-[3-(dimethylamino)propylamino]propyl}perylene-3,4,9,10-tetracarboxylic diimide (PDINN).

27. A method for preparing a perovskite/organic tandem solar cell comprising:
cleaning a transparent substrate and carrying out a UV treatment for a duration of time;
depositing a first hole transport layer on the transparent substrate;
depositing a wide-bandgap metal halide perovskite-type active layer having a bandgap greater than 1.8 eV, including a 2-amino-4,5-imidazoledicarbonitrile (AIDCN) on the first hole transport layer;
depositing an interconnecting layer on the wide-bandgap metal halide perovskite-type active layer;
depositing an organic donor-acceptor complex material as an active layer on the interconnecting layer; and
depositing a second electron transport layer on the organic donor-acceptor complex material.

28. The method according to claim 27, wherein the wide-bandgap metal halide perovskite-type active layer including the 2-amino-4,5-imidazoledicarbonitrile (AIDCN) is prepared by adding the 2-amino-4,5-imidazoledicarbonitrile (AIDCN) with a concentration of 1.5mg/mL into a perovskite precursor solution comprising 40% to 50% of bromide component; and performing spin coating to obtain the wide-bandgap metal halide perovskite-type active layer

with a bandgap greater than 1.8 eV and with the 2-amino-4,5-imidazoledicarbonitrile (AIDCN).

29. The method according to claim 27, wherein the step of depositing the interconnecting layer on the wide-bandgap metal halide perovskite-type active layer comprises:

depositing a first electron transport layer on the wide-bandgap metal halide perovskite-type active layer;

sputtering a transport conductive oxide layer on the first electron layer; and

thermally evaporating a second hole transport layer on the transport conductive oxide layer.

30. The method according to claim 27, further comprising depositing silver particles as an electrode on the second electron transport layer.