Development of Stable and Optimized Bandgap Halide-Chalcogenide Perovskite Materials for Photovoltaic Applications

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ABSTRACT

The toxicity and instability of some metal-halides has made it unfovarouble to be used for the development and fabrications of perovskites despite its good Power Conversion Efficiencies (PCE). The need to suggest good perovskites absorber materials that has excellent Dimensionality, optical band gap, and power conversion efficiencies to substitute lead in photovoltaic application is very essential in the recent research. This research work reviewed and suggested the combinable good perovskites absorber materials that can substitute lead in the perovskites. This research further discussed the possible alternative elements to replace lead in metal halide perovskites and the properties of the corresponding perovskite materials based on recent theoretical and experimental studies. In this research work other elements such as germanium, copper, antimony, bismuth, and the corresponding perovskite compounds are already showing promising properties.

Keywords: Perovskites, optimized bandgap, photovoltaic applications, Solar Photovoltaic Fabrications, Metal-halides, lead.

INTRODUCTION

Energy from the sun can be considered as the main source of all types of energy. There are several generations of PV technology which perovskite is one of the most recent researching generations that is gaining ground. Solar PV technology is one of the renewable technologies which have a potential to shape a clean, reliable. scalable and affordable electricity system for the future. Perovskite cells are leadhalide base with crystal structures[1][2]. The performance of solar cells based on hybrid halide perovskites has an unparalleled rate of progress and understanding of underlying physical chemistry of these materials trails behind [3]. The most viable area of application of perovskites materials is in buildingintegrated PV systems. The low cost, and negligible flexibility thickness requirement of perovskites materials make it an ideal for building integrated PV systems. Its high absorption coefficient, tunable bandgap. low processing temperature and abundant elemental components give it an advantage over

other thin film absorber materials [4]. Recently, the Perovskite-based solar cells have been developed and their power efficiency conversion (22.1%)has surpassed the efficiencies of other solar technologies for building-integrated PV systems such as thin-film PV cells (copperindium-gallium-selenide (CIGSe :12%). copper-indium-gallium-sulfide (CIGS). cadmium-telluride (CdTe :11%). amorphous silicon (a-Si :8%), organic PV cells (dye-sensitized (10%) and polymer (3%) [5]. A perovskites material is a hybrid solar

cell material known as organic-inorganic solar cell materials. The general chemical formula for perovskite material is given by ABX3 where A is organic or inorganic cation (Na⁺, CH₃NH₃⁺ etc), B is inorganic cation (Pb^{2+,} Sn^{2+,} Zn²⁺, Fe²⁺ etc) and X is inorganic anion (Cl⁻, Br⁻, I⁻, O²⁻). It is produced from the by-products of crude (methyl ammonium and ethvl oil ammonium) and inorganic compounds (lead iodide, tin iodide, zin oxide). Recently, organic-inorganic hybrid

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perovskites have attracted attention as light harvesting materials in mesoscopic cells [6]. Perovskite materials have been extensively used as light-absorbing material in solar cells because of their unique properties such as direct optical band gap, broadband light absorption, bipolar transport, long carrier diffusion length and flexibility [7].

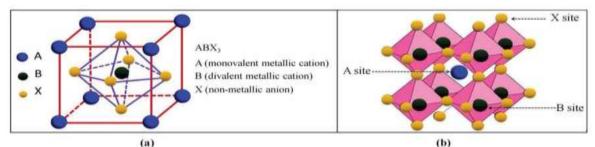


Figure 1: a) Schematic view of cubic perovskite crystal structure for ABX3 compound, b) 3D crystal structure in which the A site is confined within a cage determined by the octahedral coordination of B site with X site [8]

Figure 1, 'a' and 'b' above showed the schematic crystal structure and 3D crystal structure of Perovskites respectively. Figure one showed the crystal structure of the perovskites with a formula of ABX3 whereas A is organic or inorganic cation $(Na^+, CH_3NH_3^+ \text{ etc})$, B is inorganic cation $(Pb^{2+}, Sn^{2+}, Zn^{2+}, Fe^{2+} \text{ etc})$ and X is inorganic anion (Cl^-, Br^-, I^-, O^2) .

Perovskite solar cells hold an advantage over traditional silicon solar cells in the simplicity of their processing. Traditional silicon PV cells require expensive, multiprocesses, conducted step at high temperatures (>1000 °C) in a high vacuum in special clean room facilities. Meanwhile, the organic-inorganic perovskites material manufactured with simple is wet chemistry techniques in a traditional laboratory environment. The raw material is readily available from crude oil deposits and inorganic compound deposits [9]-[17]. The high efficiencies and the very low production costs of perovskite solar cells have made it commercially attractive to investors. Because of the high absorption coefficient, a thickness of only about 500 nm is needed to absorb solar energy. Despite the high conversion efficiency of perovskites material, current perovskites materials are unstable in the presence of thermal stress and moisture [18]. Due to the negligible thickness of perovskite solar cells, it degrades quickly in moist environments[19].

The perovskites material uses lead, tin halide-based material or transition metal based as the light-harvesting active layer. At present, most common perovskites material is lead-based material. Hybrid organic-inorganic lead-based perovskites represent a class of light absorbing materials that has led to record-breaking solar conversion efficiencies. The conversion efficiency of lead-based perovskites materials has increased from 3.8% in 2009, 22.1% in early 2016 and 24.6% in 2022, making it the fastestadvancing solar technology [4] [18] [20]. The high efficiencies of lead-based perovskites have mainly been achieved using methyl ammonium lead-halide based perovskites such as methyl ammonium lead (II)Iodide $(CH_3NH_3PbI_3)$ and (CH₃NH₃PbI₃-xClx). The CH₃NH₃PbI₃ has diffusion length of 100nm while the CH₃NH₃PbI₃-xClx has diffusion length of 1μm. The application of thermal engineering in FAPbI3 perovskite material via radiative thermal annealing and in situ XRD, effectively and efficiently monitors temperature-induced the phase transformation dynamics. It also monitors the crystal structural transformation and degradation in FAPbI₃ film but has poor production rate, rapid ramp rates for processing with low throughput [21]. In [22] an approach to further enhance solar spectrum utilization is the graded band gap, but this has not been previously

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achieved for perovskites. It demonstrated graded band gap perovskite solar cells with steady-state conversion efficiencies averaging 18.4%, with a best of 21.7%, high fill factors of ~75% and short circuit current densities up to 42.1 mA/cm², all without reflective coatings. These cells, which are based on a novel architecture of two perovskite layers (MASnI₃ and MAPbI₃xBrx), incorporating GaN, monolayer hexagonal boron nitride, and graphene aerogel, display the highest efficiency ever reported for perovskite solar cells but still have the shortcoming of toxic effect of the lead [22]. However, the poisonous nature of lead has made further research in leadbased perovskites materials unattractive [5].

The attempt to replace toxic lead-based perovskites using non-toxic tin-based perovskites has met challenges. The drawback of tin-based perovskites was its low conversion efficiency and it degrades faster compared to lead-based perovskites [23][24]. To overcome the drawbacks of group (IV) elements such as instability, low efficiency and its toxic nature, the new research is focused on improving the fabrication process when using tin or making use of transition metal-based perovskites materials. In the work done in [25], copper, silver and gold were proposed to replace the toxic lead component of current perovskites material. In [26] and [24], an alternative method to obtain lead free halides was introduced and it is based on semiconducting (hybrid) lead halide perovskites structure, where monovalent organic cation are methyl ammonium $(CH_3NH_3^+,$ MA+), formamidinium $(CH(NH2)2^+, FA^+)$ or an inorganic cation (K⁺, Rb^+ , Cs^+), divalent Pb^{2+} metal cation and the X-site of the perovskite structure is occupied by halide counterions (Cl⁻, Br⁻, I⁻). These elements were selected because they are the best conductors in existence. Other transition materials can also be used to replace the lead

The properties of lead perovskites can be tuned by changing A-site or X-site ions and also mixed ion approaches turned out to be beneficial for the performance of the perovskites in photovoltaic devices.

This project will be nanotechnology based which will focus on the development of alternative Perovskite materials composed of non-toxic elements, probable nonpresenting carcinogenicity and/or improved stability. Key challenges will involve fabricating new Perovskite materials, incorporating the materials in photovoltaic solar cells. and characterization of device's performance.

Limitations of the existing Photovoltaic **Fabrications and Optimizations**

The existing Perovskite materials for photovoltaic applications have some limitations which I want to overcome in this research. Some of the limitations are:

- > Tin-based perovskites material has low conversion efficiency and it is unstable in the moist and hot environment. These two properties make it unsuitable for buildingintegrated PV systems.
- Lead-based perovskites material is toxic and has probable carcinogenicity, thus, unstable in the moist and hot environment. These three properties make it unsuitable for building-integrated PV systems.
- Existing transition metals-based Perovskite materials have low conversion efficiency.

Aim of the Study

This research work basically aimed at:

- Proposing some Perovskite materials that will overcome the problems of existing toxic. probable carcinogenicity and/or stability problems of existing Perovskite materials.
- Suggesting the suitable way of characterizing and fabricating a new formed material.

Materials that can Substitute lead in Perovskites

Tin-based Perovskites (MASnX₃) Tin-based perovskites (MASnX₃) according to research has revealed a direct bandgap of 1.20-1.35 eV, electron mobility of 2320 cm2 V-1 s-1, hole carrier mobility of 322 cm² V-1 s-1, and long charge carrier diffusion length of more than 500 nm. The alteration of Br-/I- ratio in MASnI^{3-x} Brx has resulted in large value of VOC (0.88 V) in $MASnBr_{\scriptscriptstyle 3}$ and JSC (12.33 mA $cm^{\scriptscriptstyle -2})$ in MASnIBr₂. The absorption band can be tuned by altering the composition of halide anions in MASnX3 perovskites. However, Sn-based perovskites suffer from degradation in air due to oxidation of Sn²⁺ into Sn^{4+.} The incorporation of additives has resulted in reduced oxidation and better stability in air. The A-site cation has significant effect on photovoltaic а performance. The use of diethylammonium (en) and FA+ at the Asite of ASnX₃ has resulted in wider bandgaps and improved stability. An efficiency of 8% has been achieved for $(FA)_{0.75}(MA)_{0.25}$ SnI₃ with a VOC (0.61 V) and bandgap (1.33 eV) [8], [27], [28].

Germanium-based perovskites (AGeI₃)

Germanium-based perovskites has an optical bandgap of 1.63 eV, excellent hole and electron conducting behavior, and better stability in air. Using DFT calculations, it has been reported that with increase of size of halide anion, the bandgaps have decreasing values of 3.7. 2.81, and 1.61 eV, respectively. The replacement of the iodide content in AGeI₃ bv bromide results in enhanced photovoltaic performance and stability to a slight extent. Mixed Ge-based perovskite $RbSn_{0.5}$ Ge_{0.5} I₃ exhibits a better optical absorption and effective masses for higher carrier mobility and good stability in water. By engineering the size of A-site cation, its doping with another suitable cation and size of halide anion, it is possible to fabricate a Ge-based perovskite as an efficient light absorber [8], [27], [28].

Bismuth-based perovskite (MA)₃Bi₂I₉

The bismuth-based perovskite (MA)₃Bi₂I₉ has displayed low values of solar power conversion efficiencies up to 1.64 eV up till now, yet they have exhibited excellent stability in ambient air at room temperature and against exposure to humidity .The morphology of (MA)₃Bi₂I₉ films can be enhanced by addition of various concentration of NMP into the precursor solution that not only controlled the rate of crystallization but also enhanced the efficiency and stability in a humidity of 50-60%. relative The concentration of perovskite solution and substrate temperature also impacts its efficiency and stability. The wide bandgaps of lead-free perovskites can be engineered to a narrow bandgap by incorporating triiodide into P(4-methyl piperidinium)₃Bi₂I₉((MA)₃Bi₂I₉) that exhibited a bandgap of 1.58 eV in comparison to 1.5 eV of MAPbI₃. The various deposition methods have a direct influence on morphology of films. Cs₃Bi₂I₉ has displayed an efficiency of 8% with a pure crystalline phase and stability[8], [27], [28][12], [13].

Bismuth-based double perovskite (Cs₂AgBiBr₆)

Bismuth-based double perovskite like Cs₂AgBiBr₆ exhibited an indirect bandgap of 2.19 eV. The DFT calculations have further revealed that the family of 3D double perovskites have optical bandgap in the visible range and low carrier **Bismuth-based** effective masses. perovskites can be thoroughly investigated for enhancement in their efficiency as these materials have excellent stability in ambient air and in relative humidity. In antimony-based perovskites, the size of cationic or anionic species and the employed processing technique determines the structure. When A-site cation Cs+ is replaced by a smaller cation Rb+, a 2D layered phase is achieved with a formation energy difference of 0.25 eV comparison to Cs-based in counterparts. The addition of additive in 0D (MA) 3Sb2I9 films has resulted in enhanced light absorption in the visible

wavelength regions up to 400 nm. The use of chalcogenide and mixed perovskite materials can be an effective strategy for formation of efficient, cheap, and stable solar cells. Cs₄CuSb₂C₁₂, besides having photo and thermal stability and resistance to humidity, have exhibited excellent photovoltaic properties. There has been a significant effect on photovoltaic parameters on substitution of Sb with Bi in perovskites 2Dmixed lavered (NH4)3(Sb1-xBix)219. Bv proper substitution of Bi into antimony-based perovskites, it is possible to fabricate light harvesters with high efficiency and stability [12], [13].

Copper-based perovskites ((MA)₂CuCl₂Br₂)

Copper-based perovskites usually form 2D layered structure owing to their smaller ionic radii. By proper tuning of Cl-/Brratio, the optical absorption of Cu-based perovskites can be extended in the nearinfrared region. The (MA)₂CuCl₂Br₂ and $(MA)_2CuCl_{0.5}Br_{3.5}$ have reported better stability under ambient conditions. The divalent cations of alkaline earth metals like Mg²⁺, Ca²⁺, and Sr²⁺ can be effective replacement for lead. The perovskite solar cells based on transition metals Ti. Ni. and Cd-doped BiFeO₃ as a light absorber have displayed VOC values of 0.77, 0.56, and 0.49 V, respectively. By suitable selection of A and B-site cations and halide anions, their alteration in composition and synthesis method, it is possible to fabricate lead-free perovskites with maximum efficiency and stability without any toxic influence on environment [12], [13], [16].

Antimony-based perovskites

halide perovskites Antimony are а alternative to lead-based potential semiconductors perovskite for photovoltaic applications to address the chemical stability and the toxicity issue. The trivalent Sb^{3+} metal cation (1) is isoelectronic to Sn^{2+} (4d¹⁰ 5s²) and has a similar s² valence electronic configuration as Pb^{2+} (5s² lone pair), (2) has a comparable electronegativity (Sb: 2.05, Sn: 1.96, Pb: 2.33) but (3) a significant smaller ionic radius (76 pm) compared to the divalent Sn^{2+} (110 pm) and Pb^{2+} (119 pm) metal

cations. Because of the difference in the oxidation antimonv state. halide perovskites have the basic formula $A_3Sb_2X_9$ (X = Cl, Br, I), where A are organic (e.g. NH4⁺. $CH_3NH_3^+$, dimethylammonium, trimethylammonium,tetramethylammoniu m, guanidinium) or inorganic (e.g. Rb⁺, Cs⁺) cations. The structural chemistry and dimensionality of antimonv halide perovskites are significantly influenced by the choice of cationic and anionic species. Depending on the dimensionality, the crystal structures of antimony-based perovskites featuring Sb₂X₉³⁻ enneahalide ions within the anionic sublattice can be divided into (a) zero-dimensional, isolated double octahedral structures comprising pairs of face-sharing SbX₆ octahedra, which form discrete complex anionic metal halide Sb₂X₉³⁻ clusters arranged in dimer units. (b) infinite one-dimensional double chains of corner-sharing SbX₆ octahedra forming zigzag-type polyanionic Sb₂X₉³⁻ sublattices (c) two-dimensional corrugated double-layered polyanionic structures based on corner-connected SbX₆ octahedra to $Sb_2X_9^{3}$ [27][29-33].

Methods to adopt using different metals and solvents.

The following approaches are suggested to be adopted by researchers and Photovoltaic developer using a suitable metal:

- 1. Selection of the appropriate solvent such as Lewis acid reagent, Lewis base reagent and adducts activator Lewis base.
- 2. Selection of homo and heterovalent metal that can characteristically substitute lead
- 3. Integrating the developed and/or selected material into photovoltaic system using appropriate techniques.
- 4. The developer has to be characterized using state-of-art equipment available
- 5. The data has to be collected experimentally from the laboratory and finally tested.

Summarily, these steps and concepts that were setup in this research paper will be of a great help to the researchers and solar

photovoltaic developers as it has the working prototypes that can be used for a

This research work exposes the combinable metals and solvents that can be combined to develop a perovskites material that are free from toxicity, soluble when exposed to the air, low bandgap and low efficiency photovoltaics. This research work detailed the advantages and the disadvantages of using some of the metals and finally recommend some metals to be

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successful development and fabrications of perovskites photovoltaics.

CONCLUSIONtheused by solar photovoltaic developerscandepending on what the specifications ofitesdeveloper or what he wants to optimize. Italsooutlined sequential methods to beandused when developing and fabricatingrchperovskite solar photovoltaic panels fortheeffective and efficient power conversiontalsefficiency.

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