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A Review on Recent Advances in Materials of Hybrid Organic–Inorganic Perovskite Solar Cells

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Abstract

This study is an emphasis on the metal halide perovskite solar cells that are susceptible to factors that influence their power conversion efficiency (PCE). Perovskite solar cells, also known as PSCs, have been shown to have a high power conversion efficiency (PCE) due to a number of various factors. As they reached a power conversion efficiency of 25%, solar cells based on metal halide perovskite were a game-changer in the quest for photovoltaic performance. A flurry of activity in the fields of structure design, materials chemistry, process engineering, and device physics has helped the solid-state perovskite solar cell to become a leading contender for the next generation of solar energy harvesters in the world today. This follows up on the ground-breaking development of the solid-state perovskite solar cell in 2012. This cell has a higher efficiency compared to commercial silicon or other organic and inorganic solar cells, as well as a lower cost of materials and processes. However, it has the disadvantage that these high efficiencies can only be obtained with lead-based perovskites, which increases the cost of the cell. As a result of this fact, a new study area on lead-free metal halide perovskites was established, and it is now exhibiting a remarkable degree of vibrancy. This provided us with the impetus to review this burgeoning area of research and discuss possible alternative elements according to current theoretical and practical investigations that might be utilized to replace lead in metal halide perovskites as well as the features of the perovskite materials that correspond to these elements.

1. Introduction

Metal halide perovskites as absorber materials in perovskite-based solar cells are considered to be one of the most exciting developments in photovoltaic technology for the next generation of solar cells [1, 2, 3]. This is shown by the phenomenal increase in power conversion efficiency (PCE), which has gone from 3.8% in 2009 to over 22% at the present time in only a few short years [4, 5, 6]. The exceptional performance of metal halide perovskites may be attributed to their unique properties, which include high charge carrier mobilities [7], controlled electron and hole transport [8], high absorption coefficients [9, 10, 11], direct and adjustable band gaps [12, 13, 14], and long carrier diffusion lengths [15]. Metal halide perovskite semiconductors are particularly appealing due to the ease with which they can be processed at low temperatures. This is one of the many significant advantages of metal halide perovskite semiconductors. Other advantages include the fact that they can be created using a variety of processing technologies, such as solution and vacuum-based processes. High-throughput structure-based computations were performed [16, 17] on hypothetical three-dimensional halide perovskites (ABX_3) created by replacing lead with a number of other divalent cations taken from the periodic table. According to the scientists, who examined hundreds of material combinations, the only perovskites with noteworthy properties for use in optoelectronic applications are those based on the elements lead, tin, and germanium. $AgBi_2I_7$, a lead-free compound with a power conversion efficiency of 1.22 percent, has been reported in solar cells as an alternative material for lead-free perovskite in solar cell applications. A novel family of lead-free perovskite metal halides is denoted by the chemical formula A_2BCX_6, where A denotes a monovalent molecule, B and C denote metal cation groups, and X denotes a halogen anion [18]. These perovskites are characterized by their absence of lead. Researchers in the field of solar cells have shown interest in studying the effects of combining these materials [10, 13, 19, 20]. As a direct result of this, a variety of materials based on have developed as lead-free alternatives for use in solar cell filters. Studies [21–32] have been conducted on antimony-based perovskite materials for use in solar applications. Some examples of these are $Cs_2SbCuCl_6$, $Cs_4SbCuCl_{12}$, $Cs_2SbAgCl_6$, Cs_2SbAgI_6, and $Cs_2SbAgBr_6$. These materials have generated a great deal of attention in solar cell applications over the course of the last decade as a result of their one-of-a-kind features, which include stability in air and high temperature as well as charge carrier conductivity. As a direct result of this, the formulae of the chemicals that make up the perovskite structure may in fact be condensed into Table (1).

Table (1). Summary of Pb-Free Perovskites and Perovskite Derivatives [21].

family	Stoichiom.	Compound	solut	Solid st	Gap (eV)	PCE(%)
Perovskite	1-1-3	$MASnI_3$	x		1.23	6.4
		$MASn\ I_{3-x}\ Br_x$		x	1.41	5.73
Vacancy-ordered double perovskite	2-1-6	Cs_2SnI_6	x		1.26	6.94
		Cs_2SnBr_6	x		2.7	0.04
		Cs_2SnCl_6	x		3.9	0.07
Two- dimensional perovskite	3-2-9	$Cs_3Bi_2I_9$	x		2.2	1.09
		$MA_3Sb_2I_9$	x		2.4	0.5
		$MA_3Bi_2I_9$	x		2.1	0.12
		$MA_3Bi_2I_9$	x			0.2
		$Rb_3Sb_2I_9$	x		2.1-2.24	0.66
Double perovskite (elpasolite)	2-1-1-6	$Cs_2CuSbCl_6$	x		1.5	0.94
Other perovskite	1-2-7	$AgBi_2I_7$	x		1.87	1.22
Other perovskite	1-1-5	$HDABiI_5$	x		2.1	0.027

Table (1) is a summary of the recently disclosed lead-free materials, as well as the compounds and derivatives of perovskite materials, as well as the efficiency of the manufactured photovoltaic devices, and it also mentions the methods for preparing lead-free perovskite compounds. Table (1) can be found here. In addition to the 2D perovskites that were discussed before, 2D double perovskite halides were developed by attempting to insert organic elastomeric cations into normal 3D double perovskites. These cations include propylammonium (PA), octylammonium (OCA), and 1,4-butyldiammonium (BDA) [33]. This article survey recent advances in perovskite cells prepared from different materials, which enable them to improve their efficiency and give the best results.

2. The Perovskite Crystal Structure and Its Electronic Properties

Perovskite materials have attracted a lot of attention because to the distinctive thermal, electromagnetic, and optical capabilities that they possess, as well as the cubic lattice-nested octahedral layered structures that they have [34]. The structure of perovskite may go through a few different stages [35]: 1). The orthorhombic structural phase" at a temperature of 160 degrees Kelvin. 2) The tetragonal structure enters the phase "" when the temperature falls below 330 degrees Kelvin. 3) The cubic structure phase "" is reached when the temperature is 330 degrees Kelvin [36]. There are two primary types of crystal structures that may be found in perovskite: 1) halo-alkanes perovskite, and 2) organic/inorganic halide perovskites " [35], both of which have the ABX_3 structure [7]. As can be seen in Figure (1), the term "perovskite" refers to any substance that has a crystal structure that is comparable to that of the mineral $CaTiO_3$, what was found in 1839 [35] seen in Figure (1) and is known as perovskite [31].

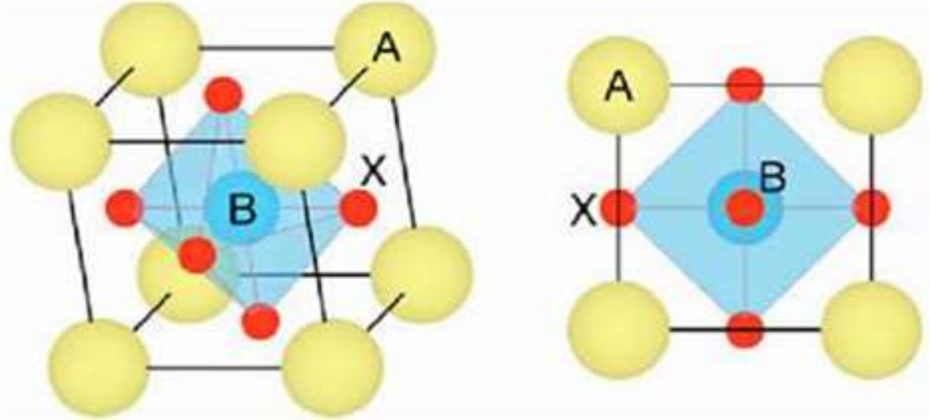

Figure (1). A general crystal structure of perovskite with the formula ABX_3 [31].

In the ABX_3 formula, A is a big cation that may be either organic or inorganic, and it is positioned the face-centered cubic lattice vertex. [35]. Examples of large cations are formamidinium (FA^+, $CH(NH_2)^{2+}$) or methylammonium (MA^+, $CH_3NH_3^+$). Group B is a minor inorganic cation, and examples of it include Cu^{2+}, Sn^{2+}, and Pb^{2+}. X_3Cl^-, Br^-, and I^- are all instances of an ion that may bind with both A and B, as seen in Figure (2).

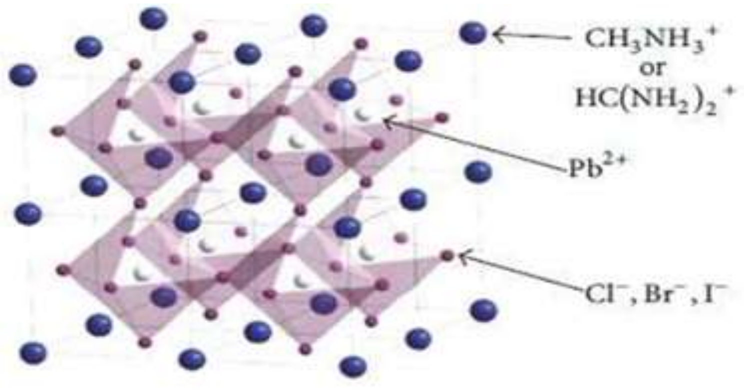

Figure (2). Cubic lattice of perovskite structures [34].

In the last several years, lead halide perovskite solar cells, also known as LHPSCs ($CH_3NH_3PbX_3$, with X = Cl, Br, and I), have attracted a lot of interest due to the fact that their manufacturing processes are more straightforward and they are less expensive than standard silicon sun cells [37, 38]. Particularly, lead-iodine perovskite ($CH_3NH_3PbI_3$) with a direct band gap of 1.5 eV has been high extinction coefficient, low exciton binding energy, excellent charge carrier mobility, simple processing capabilities, and inexpensive cost make it one of the most appropriate light absorbers. All of these characteristics make it an excellent candidate for use in light absorption. PCE has already surpassed the 25% mark at this point [39], although it is anticipated that future advancements in technology would result in a rise thanks to the materials and technologies involved, $MAPbI_3$-xClx chloride/iodide mixed halide perovskite is employed in a broad range as a result of the thermal stability of the $MAPbI_3$-xClx films, which is up to 140 degrees Celsius, as well as its superior diffusion lengths greater than $MAPbI_3$ (Iodide of methyl-ammonium lead) [36].

Despite the significant achievements of perovskite solar cells, the most significant barrier to the widespread deployment of PSCs continues to be lead's toxicity [40]. As a consequence of this, the instability and toxicity of lead PSCs may limit both the scale at which they may be produced commercially and their possible applications. Because of these issues, there is a pressing need to look for a different potential ecological hybrid perovskite material that would have the same level of effectiveness [24]. Tin halide perovskite solar cell $CH_3NH_3SnI_3$ ($MASnI_3$) with a narrow bandgap of 1.3 eV has been shown through both experimental and theoretical research to have the potential to serve as a suitable replacement for LHPSCs. This is because $MASnI_3$ covers a greater portion of the visible light spectrum than LHPSCs do, which is the reason for this result. In addition, a planar heterostructure made of Tin (Sn) PSCs has been built, and it has achieved an efficiency of 3 percent while having an optical bandgap that spans from 1.3 eV to 1.4 eV. This was accomplished by constructing the heterostructure on a flat surface. In addition, the bandgap may be adjusted in a manner similar to that of LHPSCs from 1.3 eV to 2.15 eV by swapping iodine (I) for bromine (Br), which results in a rise in the bandgap value. This causes the bandgap value to increase from 1.3 eV to 2.15 eV. This bandgap tunability gives tin-based perovskites a high opportunity to be used in solar cell applications as a viable replacement for LHPSCs [22], [28]. The oxidation of tin from Sn^{2+} to Sn^{4+} in the air is the primary factor that prevents the performance of tin-based perovskites from reaching its full potential [31, 33, 34]. Extensive research has led to the development of encapsulation methods, which in turn has increased the stability of Sn-based devices. Tin fluoride (SnF_2) is added to the synthesis process in order to reduce the likelihood of oxidation from stannous two plus to stannous four plus [31, 33]. Due to the fact that it is in the same subgroup as lead, the perovskite based on germanium (Ge) ($CH_3NH_3GeX_3$) exhibits the same level of performance as lead (Pb) and tin (Sn). The 2013 was the year when Stoumpos et al. made history by being the first individuals to successfully synthesize the lead-free germanium iodide perovskite materials $CH_3NH_3Gel_3$. They also made the discovery of a highly distorted structure as well as powerful nonlinear optical characteristics. As a direct result of this, Krishnamoorthy et al. in 2015 developed $CH_3NH_3Gel_3$, which has a significant amount of promise in photovoltaic applications. A mixed Ge–Sn-based perovskite material was recently synthesized by Nagane et al. (2018). This material is appropriate for use in more efficient single-junction SCs [24].

3. Materials for the Structure and Photovoltaic Devices

These factors bring up an essential question: is it feasible to totally substitute Pb in perovskites solar cells with benign materials without losing efficiency and stability, or is Pb absolutely required for the process? Two recent computer investigations made an effort to find an answer to this issue by searching through all of the various hypothetical halide perovskites that may be produced by the monovalent substitution of lead. The authors of reference 40 employed the low-temperature orthorhombic phase of $CH_3NH_3PbI_3$ as a starting template to examine the electronic structure of perovskites having the formula $CsB^{2+}X_3$, where X = Cl, Br, or I, are hypothetical. They did this in order to better understand how these perovskites would behave. The authors took into account every conceivable element that may potentially produce di-halide salts for the metal cations B^2+, which resulted in the discovery of more than a hundred different compounds. In an effort to stabilize Sn-based perovskites, a number of scientists halide perovskite compounds based on Sn that have been reported to have the same overall composition A_2SnX_6, such as Cs_2SnI_6, $Cs_2Sn(I, Br)_6$, and Cs_2SnX_6 with X = Cl, Br, and I. These derivatives have the general composition A_2SnX_6. Due to the presence of tin in its stable +4 oxidation state, these compounds maintain a high degree of stability when exposed to air and water, which contributes to their composition. Cubic double perovskites $Cs_2Sn_2X_6$ are created by eliminating half of the Sn atoms from the octahedral Sn atoms., which results in a molecular salt that is made of Cs^+ cations and $[SnI_6]_2$ octahedral as anions, as shown in Figure (3). Due to the fact that the Sn vacancies in the double perovskite structure establish a regular sub-lattice, these compounds are referred to as double perovskites, and their formula is designated by the letters Cs_2SnI_6 [24].

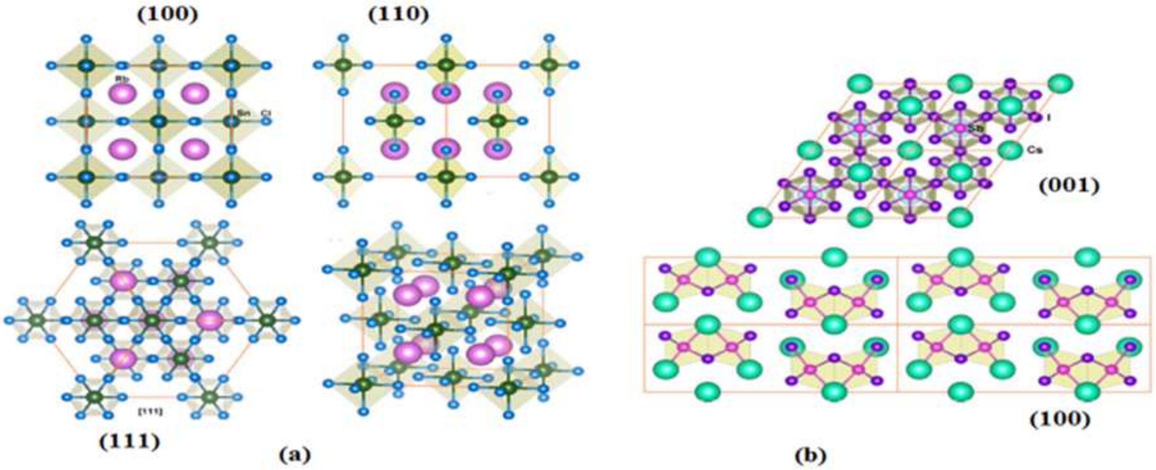

Figure (3). Structure models of Cs_2SnI_6 with 0-dimensional perovskite structures, observed along [100], [110], [111], and perspective view of the structure. (b). Structure models of $Rb_3Sb_2I_9$ with 0-dimensional double perovskite structures, observed along [001], and [100] [24].

These compounds may be conceived of as $A_3B_2X_3$ perovskites; however, in order to preserve charge neutrality, one out of every three octahedral B^{3+} sites are left empty in each of them. Crystallization takes place in the P3m1 space group for the structures that are created, which are referred to as "two-dimensional layered perovskite derivatives as shown in Figure (4). In these compounds, the cation Pb^{2+} is substituted by either Sb^{3+} or Bi^{3+}, such as in $Cs_3Sb_2I_9$ and $Rb_3Sb_2I_9$. When significant A-site cations are present, these structures have the potential to convert into zero-dimensional face-sharing BX_6 octahedra belonging to the space group P63/mmc, as can be seen in $MA_3Sb_2I_9$ and $MA_3Bi_2I_9$. All of these compounds exhibit band gaps in the vicinity of 2.1 eV, and their stability in air is superior to that of $MAPbI_3$. There have been reports of solar cells made using $Cs_3Bi_2I_9$ (1.09 percent PCE), $Rb_3Sb_2I_9$ (PCE = 0.66 percent), and $MA_3Bi_2I_9$ (PCE = 0.2 percent), as well as planar hetero-junction devices made with $MA_3Sb_2I_9$ (PCE = 0.5 percent). Pb-free double perovskite halide compounds have been found for the following: $Cs_2AgBiBr_6$, $Cs_2AgInCl_6$, $Cs2CuSbCl_6$, $Cs_2NaBiCl_6$, Cs_2KEuCl_6, $Cs_2LiScCl_6$, $(CH_3NH_3)_2AgBiBr_6$, $(CH_3NH_3)_2KBiCl_6$, $Rb_2NaCrCl_6$, and others There is a possibility that some of these double perovskite elpasolite chemicals might be used in lead-free solar cells [10], in addition, reports have been made about the energy deficits [13]. It is anticipated that double perovskite elpasolites will also be used in thermal neutron scintillator material applications [22, 25]. As can be seen in Figure (5), each of the BX_6 octahedra is located on its own inside the perovskite crystal, making it a 0-dimensional (0D) perovskite. Because the combination of $Cs_2AgBiBr_6$ in Figure (4a) does not include enough Ag atoms to generate $Cs_2AgBiBr_6$, the $BiBr_6$ octahedra are separated from one another in the crystal, and A site cations fill the cub-octahedral gaps. The bismuth dpoint of double perovskites serves as the starting point for the incorporation of tetravalent cations into $4^+/0$ double perovskites. For vacancy-ordered double perovskites, the usual formula is A_2BvX_6, where v stands for empty locations corresponding to the B' site for the $A_2BB'X_6$ double, as shown in Figure (5). There have been reports of structural models for Cs_2SnI_6 including 0-dimensional perovskite structures. Despite the presence of solitary octahedral BX_6 units, the close-packed iodide lattice permits electrical dispersion. Cs_2SnI_6 and other perovskites have been used in the fabrication of solar cells [22]. There have been publications on solar cells that do not include lead, such as $FA_4GeSbCl_{12}$ [27], in which the double elements were selected to replace Pb.

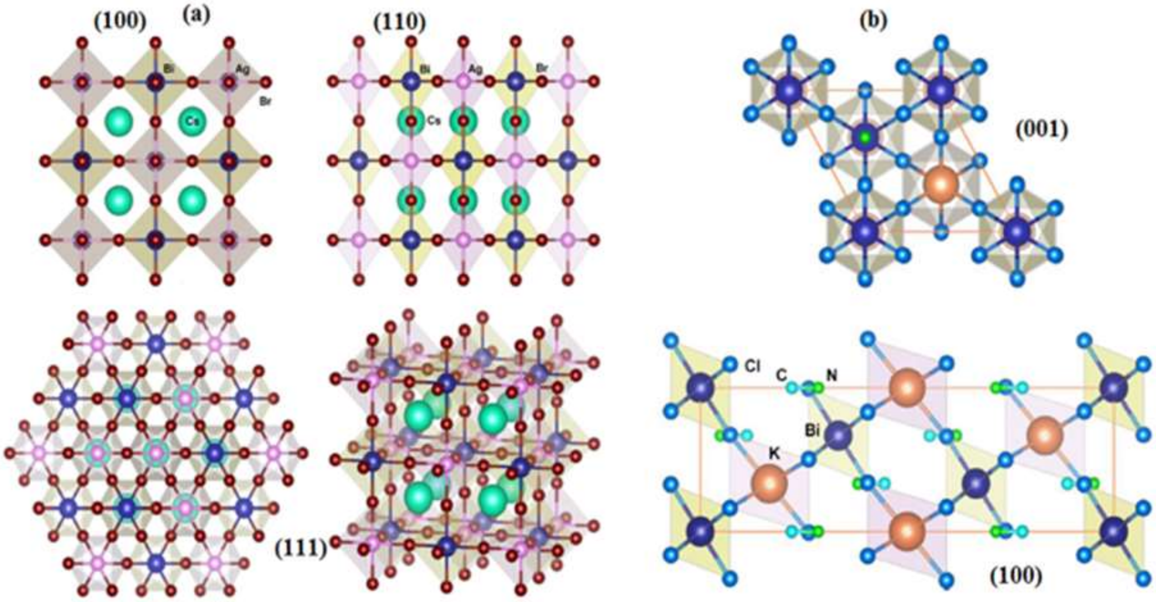

Figure (4). Structure models of (a). $Cs_2AgBiBr_6$ with double perovskite structures, observed along [100], [110], [111], and perspective view of the structure. (b). Structure models of $(CH_3NH_3)_2KBiCl_6$ double perovskite structure observed along [001], and [100] [24].

$Cs_2TiI_xBr_{6-x}$ vacancy-ordered double perovskite compounds have also been reported with bandgaps between 1.0 and 1.8 eV [23].

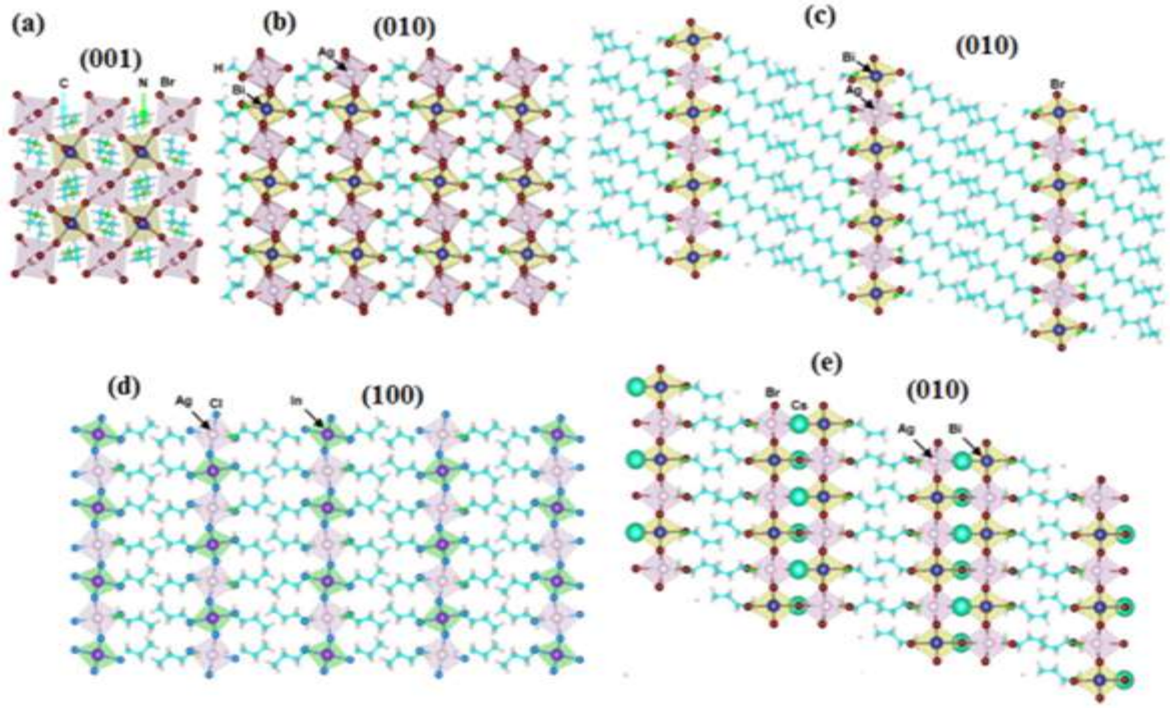

Figure (5). Structural models of (a, b) $(NH_3(CH_2)_4NH_3)_2AgBiBr_6$, (d) $(CH_2)_2NH_3)4AgInCl8$, and (e) $(CH_3(CH_2)NH_3)_2$ with 2- dimensional double Perovskite structures [24].

Recent research has resulted in the development of three distinct but related architectures for perovskite solar cells (PSCs): There are three types of perovskite solar cells: mesoporous (n–i–p), planar (n–i–p), and planar (p–i–n). All three of these PSCs are planar [41] as shown in Table (2).

Table (2). Type of structures of Perovskite Solar Cells.

Mesoporous Perovskite Solar Cell (n-i-p)	**Planar Perovskite Solar Cell (n–i–p)**	**Planar Perovskite Solar Cell (p–i–n)**
Mesoporous perovskite solar cells (MPSCs) have been getting a lot of interest as of late due to the fact that they are simple to manufacture, don't need a lot of expensive components, and have a high power conversion efficiency (PCE). The compact layer is typically formed on top of a fluorine-doped tin oxide (FTO) layer during the MPSC process [35]. FTO layers are known to block holes and extract electrons. Nanopores in an ETL scaffold are a characteristic feature of a typical MPSC [36]. First, the absorber layer completely envelops the ETL, creating a dense capping layer. Next, the absorber layer penetrates the ETL, resulting in the formation of an intermixed layer. After the absorber layer has been deposited, the device is finished by depositing the top electrode and the HTL in the appropriate order [34]. As a result, the manufacturing process must include an additional step for the mesoporous layer, which is something that should be avoided when scaling up. There is a continuing debate on whether or not a mesoporous layer is essential for enhancing the material's stability or achieving a higher PCE [37].	Planar perovskite solar cells, also known as PPSCs, have been the subject of much research for a variety of reasons, including their low-temperature manufacturing technique, cheap cost, and ease of processing [35]. A compact ETM layer is present in the planar n–i–p structure; this layer is distinct from the intermixed layer (perovskite– ETM) that is present in the mesoporous architecture [34]. There are two interfaces: one between ETL and perovskite, and another between perovskite and HTL. As a result, the e-h pairings may be efficiently and quickly separated using ETL and HTL [1].	PSCs with inverted structures (p–i–n) have a great potential for highly performed and flexible PV devices due to their numerous advantages, including simple processing techniques, high stability, and negligible hysteresis [35]. This is due to the fact that PSCs with inverted structures have a low hysteresis. Because the extraction layer of a carrier in the n–i– p structure is flipped upside down, the general term for the planar p–i–n architecture is an inverted planar structure. This is because of the inversion. It's common practice to construct a p–i–n architecture using a planar structure that has a compact HTL [34].

4. Perovskite Solar Cell Operation Principles

The solar cell is regarded as the primary component of a photovoltaic array [42]. Semiconductor materials are used in the construction of solar cells; these materials use the PV effect to turn sunlight into electrical energy [43, 44]. In order to perform its role in an acceptable manner, it must have the greatest possible PCE [45]. It is vital to have a fundamental understanding of the processes or stages that are carried out in the SC layers, as well as the role of each component, in order to build highly effective SCs and to comprehend how photovoltaic cells work [45]. When solar cells are placed in an environment where they are exposed to light, the layer of semiconductors inside the cells will absorb some of the photons that have an energy greater than Eg. The photons that are absorbed and have sufficient excitation energy ($E > E_{gap}$) cause the holes and electrons to be transported in opposite directions. This movement occurs because the holes are in the valence band and the electrons are in the conduction band, as shown in Figure (6) [43]. Because excitons in perovskites have a different binding energy than excitons in other materials, excitons in perovskites may either recombine into excitons or produce free carriers (consisting of free holes and electrons) that can generate current [46].

In the region occupied by the field, e-h pairs are first created, and then, as a result of the action of this field, they are separated. The movement of the electrons toward the n-side and the holes toward the p-side is a direct consequence of this phenomenon. In the following, certain electrons and holes will gather along the field area borders of the N-side and P-side neutralizing sections of them, respectively. The remaining electrons and holes, which are transported via the outside circuit, are responsible for serving the load [35, 47]. Following the collection of free electrons and holes concurrently by HTL and ETL, which is then followed by the collection of free electrons and holes sequentially by FTO and the metal electrode [48]. The electrostatic field may be able to negate some of the effects of the potential energy barrier, and it also has the ability to provide the P-region positive electricity while giving the N-region negative electricity.

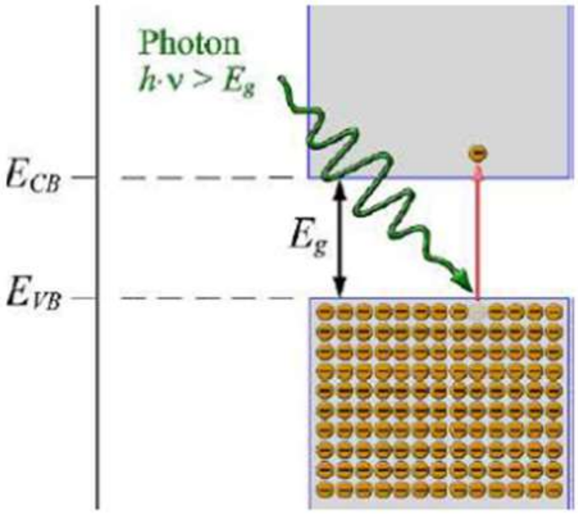

Figure (6). Photo-generation of an electron-hole pair in a semiconductor [35].

In addition to mitigating some of the negative consequences of the potential energy barrierit is possible that the electrostatic field will lead the P-region to have positive electricity while the N-region will keep its negative charge [36]. The complete process is shown in Figure (7). It has been discovered that $CH_3NH_3PbI_3$ ($MAPbI_3$) and other perovskites have longer carrier diffusion distances and lifetimes than other materials. This is because these materials have higher carrier mobility and lower carrier recombination probabilities. For instance, the distance of carrier diffusion for $MAPbI_3$ is greater than one micrometre and greater than one hundred nanometers when compared to $MAPbI_3xCl$. The excellent performance of PSCs can be attributed to the carriers' extended lifetimes as well as their greater distances of diffusion [35].

5. Fabrication Techniques

Glass/Transparent Conducting Oxide (TCO)/ ETL /Perovskite layer/HTL /Au is the composition a typical metal halide photovoltaic solar cell, also known as a PSC, has a structure that is flat and regular (n-i-p), and the composition of a PSC glass, transparent conducting oxide (TCO), high-temperature liquid perovskite (HTL), and perovskite all have an inverted planar structure, or p-in. In order to generate the mesoporous structure, an additional layer is added to the planar structure in the way that is explained further below PSC. Glass/TCO/ c-

ETL/ perovskite layer/ high temperature layer/ gold Therefore, the processes of manufacture are not uniform across all of the available options. In this article, we will conduct a brief analysis of the manufacturing processes used in earlier research for planar (regular, inverted), mesoporous, and ferroelectric PSCs.

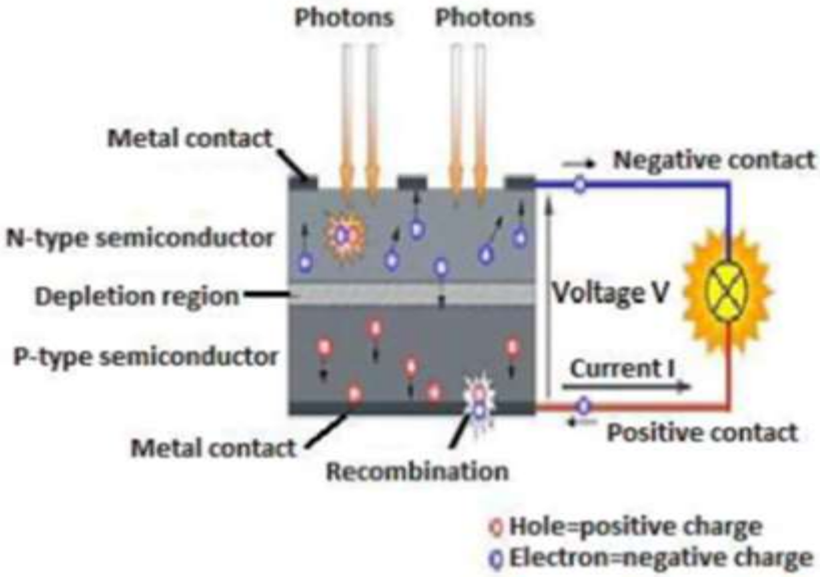

Figure (7). Solar cell working principle [36].

6. Conclusions

Perovskite solar cells have made significant strides in terms of their performance in recent years. In order to construct solar cells that have a high level of performance, several production methods and novel perovskite compounds have been developed. In recent years, a large variety of new materials, such as ETA/EDA/DTA, SZTO, MZO, $Cu_2O/MAPbI_3/SiO_2$, $TiO_2@PbTiO_3$, $CsPbBr_3$, 2D/3D Pb Sn alloys, TiO_2/SnO_2, $Li:SnO_2$, CBD-modified ZSO, PAMAM, PEDOT:PSS, etc., have been created. These new. Perovskite solar cells are the subject of major research efforts, but considerable obstacles still need to be overcome before they can be commercialized. However, in order to produce perovskite solar cells for practical applications, a large number of scientific obstacles and concerns need to be addressed. These challenges and issues include a reduction in charge separation, transportation, and collecting losses. Charge carrier injecting and collecting qualities are vital for illuminating the performance, and they are intimately related with the interfacial properties that exist between the electrodes and the photoactive layers. Among other things, this is the case. It is common knowledge that the characteristics of interface materials, such as their morphology and the alignment of their energy levels, have significant influence on the charge transfer in solar systems. These effects are crucial since they are widely recognized. Researchers are making unceasing attempts to address the obstacles, as well as to build perovskite solar cells for the next generation, which will have improved power conversion efficiency and long-term stability.

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