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ABX₃ inorganic halide perovskites for solar cells: chemical and crystal structure stability

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RESUMEN

La energía solar es una de las tecnologías más prometedoras y desarrolladas de los últimos años, debido a su alta eficiencia y bajo costo. En este sentido, las celdas solares de tipo perovskita han sido el foco de atención de la comunidad científica mundial. El objetivo principal de este trabajo es presentar un análisis de diversas investigaciones reportadas sobre la síntesis y desarrollo de películas fotoactivas de perovskita ABX₃ para celdas solares, con énfasis en el efecto que la temperatura y la humedad tienen sobre la estabilidad de su estructura química y cristalina de la perovskita inorgánica ABX₃. En cuanto a la estructura de las celdas fotovoltaicas basadas en perovskitas inorgánicas tipo ABX₃, se presenta un análisis sobre los materiales de los que están conformadas y sobre la mejora de la eficiencia (PCS), factor de llenado (FF), densidad de corriente a corto circuito (J_{sc}) y del voltaje a circuito abierto (V_{oc}) de estos dispositivos. A manera de conclusión, se presenta una relación de los métodos, variables de síntesis y tipo de perovskita inorgánica utilizados para el desarrollo de dispositivos fotovoltaicos con las mejores eficiencias; también se resaltan las tendencias hacia las que se dirige esta área importante de la ciencia.

Palabras clave: Perovskitas inorgánicas, propiedades optoelectrónicas, estabilidad química, estructura cristalina, métodos de obtención.

ABSTRACT

Solar energy is one of the most promising and developed technologies in recent years, due to its high efficiency and low cost. Perovskite-type solar cells have been the focus of attention by the world scientific community. The main objective of this article is to present an (PSCs) analysis of the various investigations reported on the development of ABX₃ inorganic halide perovskite-based solar cells, with emphasis in the effect that temperature and humidity have on their chemical and crystal structure stability. The main methods that are used to obtain ABX₃ inorganic halide perovskites are also presented and analyzed. An analysis about the structure of these photovoltaic cells and how to improve their efficiency (PCS), fill factor (FF), short circuit current density (J_{sc}) and open circuit voltage (V_{oc}) of these devices is presented. As a conclusion, a relationship of the methods, synthesis variables, and type of inorganic halide perovskite used for the development of devices with the best efficiencies is presented; the trends towards which this area of science is heading are also highlighted.

Keywords: Inorganic perovskites, opto-electronics properties, chemical stability, crystal structure, obtention methods.

1. INTRODUCTION

Solar energy is one of the most promising alternatives to handle world energy demand, for which it is necessary to develop new materials and devices, such as solar cells, that allow it to be used more efficiently. The materials used for the manufacture of photovoltaic cells are those that convert photons into electrons through the photoelectric effect. This is why the search for materials that have the ability to absorbing light for application in photovoltaic devices has focused on materials ranging from metallic oxides, such as silicon, organic dyes, semiconductor polymers, as well as other chemical structures capable of performing this function, such as metal halide perovskites, which have a small band gap, charge mobility, low excitonic energy, long diffusion lengths, and high carrier mobility. Those materials are the basis for different generations of solar cells, the latter being used in perovskite solar cells (PSCs) [1-7].

The work describing the use of perovskites as an absorbent layer was published in 2009 when Miyasaka et al., used lead halides and methyl ammonium (MA) perovskites in a Grätzel-type cell, which use liquid electrolytes. Although the efficiency of these cells was only 3.8%, they paved the way for the development of current PSCs, which have reached efficiencies greater than 22%, for which the liquid electrolyte was changed for semiconductors carrying solid hollows [8, 9]. Nowadays, the PSCs are cells n-i-p type where (n) is an electron transporting layer (ETL) typically TiO₂, (i) is the perovskite totally inorganic or with an inorganic cation and (p) is an organic or inorganic hole transporting layer (HTL) [10-12]. Although the efficiency of these devices has reached levels that makes them a real alternative for their application and use in large-scale clean energy generation, they still present points of improvement, such as the chemical and structural stability of perovskite. As mentioned before, the first perovskite used for this purpose consisted of methyl ammonium and lead halides, particularly CH₃NH₂PbI₃ (MAPI) and CH₅N₂PbBr₃ (FAPBr), which are still being used and are the ones with the highest efficiency. However, these structures have a low stability against to factors such as the presence of humidity and the presence of oxygen, in addition to being unstable at temperatures above 85 °C, low formation energy, etc., making it difficult to apply in environmental conditions. Due to the above, research is continuing on the synthesis of perovskites and the development of solar cells based on perovsquites that do not present these deficiencies [10, 13-16]. Inorganic perovskites appeared as a possible solution to the deficiencies on chemical stability presented by organic/inorganic perovskites, since temperature affects the crystal structure but does not degrade its components, which is vital for devices that will be exposed to solar radiation. Inorganic perovskite solar cells have lower efficiencies than organic/inorganic perovskite-based cells [17, 18].

The most used cation to replace another cation in the structure of the organic perovskite MABX₃ has been the Cs, radically increasing stability. Inorganic perovskites are not so hygroscopic and withstand high temperatures unlike their organic / inorganic counterpart, however, far from being presented as the definitive solution for application in photoconversion processes, they still need to be adapted, since they present different crystallographic arrangements and not all of them are photoactive [19-22].

The main inorganic perovskites currently used are CsPbI₃, CsPbBr₃ and CsPbI_yBr_{3-y}. The first one has a band gap that is very close to 1.73 eV, this structure presents a black coloration in its cubic phase that turns yellow once it decays to its orthorhombic form, which is inactive and more stable at room temperature, which is thermodynamically favorable, even when it is kept free of moisture and invariably occurs within a few days at most [23-26]. The perovskite CsPbBr₃ has a gap band width of nearly to 2.3 eV, in its cubic phase, this is the most stable of the inorganic perovskites at room temperature. It has been reported that photovoltaic devices manufactured with this perovskites exceed the 2000 hours of use, nevertheless, in comparison with CsPbI₃, it has lower photoconversion efficiency [27-31], that is why the inclusion of Br⁻ and I⁻ in the same perovskite ABX₃ (X=Br_{3-v}, I_v) lead to obtain a structure that presents better efficiencies than those that only have Br and better stability than I perovskites [32-34]. The inclusion in the crystalline network of cations such as K, Zn, Bi, has also been explored to modify the size of the crystallite and favor its stability under environmental conditions. Some researchers have chosen to replace the Pb of perovskite due to the toxicity of this element. The lead-free $CsSnX_3$ perovskite has very low percentages of photoconversion efficiency [35-40]. This work aims to provide a current perspective of the stability of inorganic ABX_3 perovskites used for the construction of solar cells, including the architecture of the devices, the structure of the perovskites, the synthesis and the additives used, as well as the efficiencies achieved and future trends.

2. PEROVSKITE SOLAR CELL DEVICES

The basic structure of PSCs shown in Figure 1, consists of an optically transparent conductive substrate, followed by a "n" type semiconductor layer responsible for the transport of electrons, which are produced in the perovskite layer "i" in which the photoelectric effect takes place. Attached to this is a gap transporter film of a "p" semiconductor and finally a conductive electrode such as gold, carbon or silver. However, it is not the only structure tested (nip), since type cells have been generated (pin) which are commonly called inverted PSCs, in both cases solar irradiation is carried out through the conductive glass which is commonly oxide of fluorine doped tin (FTO) or indium tin oxide (ITO) although the use of graphene thin films has also been reported. Some researchers have tried irradiating the cells from the conductive electrodes, although this has not presented a competitive photo-conversion efficiency, nevertheless it is important to know that these devices can take advantage of the diffuse radiation that hits the other side of the PSCs [41-46].





As already mentioned, the photoelectric effect is carried out on the perovskite, allowing the generation of electric current in these devices. This can be described as the promotion of an electron from the valence band of perovskites to the conduction band, followed by the mobility of electrons and holes (excitons) in opposite directions, being the generation of the exiton or electron hole pair, Equation 1, to later be separated through semiconductor films, said mobility which is expressed by Equations 2 and 3 for the electron in n-type films, while what happened in p-type films is described by Equations 4 and 5 [47, 48].

$$ABX_3 + h\upsilon = (e^- \dots h^+)ABX_{3x}$$
⁽¹⁾

Conducting hole across in ETL

$$(e^{-}\dots,h^{+})ABX_{3} = e^{-}ETL + h^{+}ABX_{3}$$
⁽²⁾

$$h^+ABX_3 = h^+(HTL)$$
(3)

Conducting hole across in HTL

$$(e^{-}...,h^{+})ABX_{3} = h^{+}(HTL) + e^{-}ABO_{3}$$
 (4)

$$e^{-}ABX_{3} = e^{-}(ETL)$$
⁽⁵⁾

In the general structure of PSCs, a pair of layers are required that can separate the positive and negative charges once the exiton has been generated, as shown in Equations 2-5. This process must be performed quickly, to avoid recombination of electrons, which in turn results in low photoconversion efficiency, these layers of diverse materials must be energetically coupled with the perovskite used to facilitate these processes [49-51], as shown in Figure 2. Load-bearing films can help improve the stability of perovskites, since when placed on the bottom and top they help to isolate the perovskite from external factors, therefore the correct choice of these films allows to extend the life time of these devices. In the case of HTL, there is a large number of possible materials for this purpose, among the most used materials are (2,2', 7,7'-tetrakis (N, N-di-p-methoxyphenylamine) -9, 9'-spirobifluorene)) (Spiro-OMeTAD) or poly (triarylamine) (PTTA). Another widely used material is Poly (3,4-ethylenedioxythiophene) -poly (styrene sulfonate) (PEDOT: PSS). With the aim of improving these films, modifications have been made to the most widely used films, such as the Spiro-OMeTAD, in which the modification has been reported changing the position of the substituents p-methoxy (-OMe) in ortho positions, goal and for, which allowed to identify, that the position for is the most efficient in terms of transport of holes [3, 41, 52-55].

A molecularly engineered hole transport material with a simple asymmetric fluorene-dithiophene (FDT) core replaced by N, N-di-p-methoxyphenylamine donor groups, may serve as HTL, other promising materials are allotropic forms of carbon as carbon nanotubes, which have been used for this purpose. On the other hand, the use of inorganic structures as hole carriers, such as NiOx, as well as Cu-doped oxides has also been investigated [56-58]. Also, a PSCs structure like Au/Spiro-OMeTAD/CsPbI₃/CsPbBr₃/TiO₂/FTO has been studied [59-62].



Figure 2: Schematic energy-level diagram of Au/Spiro-OMeTAD/CsPbI₃/CsPbBr₃/TiO₂/FTO.

It is important to emphasize that the negatively charged carrier must present a band gap capable of preventing the flow of electrons but facilitating the mobility of the holes because the negatively charged particles generated by the photoelectric effect carried out in the perovskite are subtracted dare of n-type semiconductor films, of which the most common is TiO_2 , either in its anatase, rutile or even brokite phase, with the purpose of improving ETL. The anatase phase of TiO_2 is the most used as an ETL in perovskite solar cells (DSSC) or as a photoanode in dye-sensitized solar cells (DSSC), using synthetic [63] or natural [64] dyes. with Y has been reported, which has allowed a better adjustment of energy levels to those of perovskite, thus preventing recombination. Other works on materials used as ETL have reported the use of SnO_2 or ZnO, mixtures of metallic oxides, and even the application of organic macromolecules such as fullerene, which has been used alone or decorated with Zn, and the graphene used to doping the TiO_2 . In particular, the use of graphene has been shown to stabilize the cubic phase of perovkites, increasing their useful life [49, 65-67].

The perovskite photoactive layer is made up of chemical structures represented by ABX₃ where "A" corresponds to a large cation, such as Cs⁺ or methyl ammonium, located in the center of the crystal, "B" is a cation located in the corners commonly Pb²⁺, although Sn²⁺ has also been used, while "X" corresponds to an anion, particularly Γ , Br⁻, Cl⁻, located in the center of the edges of the cubic cell [68, 69]. Ideally, perovskites have a cubic crystalline structure centered on the faces, this being the most desired for their photovoltaic applications. However, it is not the only structure that can be formed, since they can also have tetragonal, orthorhombic or rhombohedral phases. To evaluate this, the Goldschmidt's tolerance factor is used (Equation 6), which depends on the ionic radii of cations A and B and on anion X [68, 70]. In Figure 3, the cubic structure of perovskites ABX₃ and some factors of tolerance for various perovskites are shown.

$$\mathsf{t} = \frac{r_A + r_B}{\sqrt{2}(r_B + r_x)} \tag{6}$$

Where r_A and r_B are the ionic radio of the cations A and B and r_x is the ionic radius of the anion, if this number is equal to 1, the crystal structure will be face-centered cubic, if this value is higher, the structure will be hexagonal, if t is between 0.9 and 1 the crystallography will correspond to a distorted cube, while at values t between 0.7 and 0.9, the perovskite will present a tetragonal structure; on the other hand, at values t less than 0.7, the structures could be orthorhombic and rhombohedral. In other words, when the Goldschmidt tol-

erance factor is different from 1, distortions in the network increase [71-73]. In the case of perovskites for photovoltaic applications in its active phase, also called the black phase, t is required to be greater than 0.8 and less than 1. Likewise, the symmetric cubic phase is obtained when (0.9 < t > 1) or high temperatures. However, perovskites with organic cations such as FAPbI₃ (t \approx 1) or MAPbI₃ (t \approx 0.9) or CsPbI₃ (t \approx 0.8) that have a Goldschmidt's tolerance factor suitable for the black phase, present the inactive phase at room temperature, also called yellow phase [38, 71, 74-76].



Figure 3: Cubic structure of perovskite ABX₃ and tolerance factors of CsPbBr₃, CsPbI₃, CsPbI₂Br, and CsSnI₃ (Drawn with VESTA 3) [77].

As already mentioned, inorganic perovskites have better stability than those with an organic component, which when are irradiated by light generate superoxide O^{2-} , which reacts with the protonated organic component, meanwhile Cs⁺, which is the most common substituent "A", lacks protons in its structure, which reduces the effect of superoxide, increasing stability against the effects of light and oxygen, compared to organic componentes. However, for B²⁺ type substituents such as Sn²⁺, it usually oxidizes to Sn⁴⁺ and changes the properties of perovskites that contain it [13, 76, 77]. Another adverse effect of environmental conditions is humidity, since it favors the structural rearrangement of inorganic perovskites towards the formation of the δ -orthorhombic inactive phase [78-80]. According to Yihui Li et al., it is possible to maintain this CsPbI₂Br structure for more than 300 hours by keeping the humidity below 25% [22]. A variable to take into account is the temperature, since the photoactive phases are favored at high values, as shown in Figure 4, however, at room temperature the most stable phase (orthorhombic) is photovoltaically inactive [6, 81, 82].

Modification of components A, B or X has been the first route to follow to increase the chemical and structural stability of perovskites, either by the total or partial replacement of any of its components, or by doping that relaxes the structural stress. Efforts have also been focused on optimizing the processes for obtaining these structures, controlling heat treatment, grain size, and the use of both organic and inorganic additives [32, 82-88].

3. STABILITY OF INORGANIC PEROVSKITES

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Figure 4: Effect of temperature on the formation of active phases of CsPbX₃ inorganic perovskites (Drawn with VESTA 3) [75].

4. METHODS OF OBTAINING ABX₃ PEROVSKITES

The production of both organic and ABX_3 inorganic perovskites for application in solar devices must be carried out in such a way that highly crystalline structures are obtained, which homogeneously coat the surface and present the fewest possible defects, since the quality of the films perovskite has a significant effect on the stability and efficiency of devices. The desirable characteristics of these films can be achieved through control of crystallization, pre-treatments of the precursors, additives or catalysts, the technique of applying the precursors and by the applied heat treatment. In this context, there are various techniques that can be used to obtain high quality films by spin coating technique (using one or two steps) and vapor deposition technique.

4.1 One step technique

The chemical synthesis of the ABX_3 perovskites is carried out by solvating the precursors, in polar media such as Dimethylformamide (DMF) or Dimethyl sulfoxide (DMSO), as well as solvent mixtures, obtaining a precursor solution, which is used to coat a surface and generate the precursor film, using the spin-coating or deep-coating techniques. The use of anti-solvents to promote crystallization and the formation of high-quality films is common, the most commonly used being diethyl ether, toluene and chlorobenzene, followed by a heat treatment that allows the production of perovskite [91-97]. The composition of the precursor solution depends directly on the desired perovskite, if we take the perovskite of CsPbI₃ as an example, the use of CsI, PbI₂, as precursors will be required on a molar basis. While the required heat treatment will be up to 300 °C to favor the crystallization of the active phase [20, 98]. WANG et al., prepared the CsPbI₃ perovskite in a ratio of solvents (1: 4) DMF:MSO with precursors CsI and PbI₂, 1 M, by means of the spin-coating technique with an annealing temperature of 250 °C [99]. ZHU et al., reported the convenient presence of controlled pores in CsPbIBr₂ perovskites using the one-step technique, which provides better crystallinity and fewer film defects allowing cell efficiency of up to 9% [100]. On the other hand, ZHANG et al., studied 6 different anti-solvents; ethyl acetate, toluene, chlorobenzene, chloromethane, isopropanol and diethyl ether for the synthesis of CsPbIBr₂, showing the influence of these for crystallization, homogeneity and packing of the grains, as well as to restrict the formation of holes, denoting a better film formation when using diethyl ether [94]. Figure 5 illustrates the two-step technique for making perovskite films.



Figure 5: Schematic procedure of the one-step technique for obtaining perovskite films.

4.2 Two Steps Technique

The two-step technique consists in the addition of a first perovskite precursor, generally the precursor of Pb²⁺, or Sn²⁺ dissolved in DMF or DMSO, by spin-coating for its subsequent drying, followed by the addition of the Cs precursor dissolved in methanol or in another polar solvent, to increase the concentration of said precursor, due to the low solubility that salts like CsBr present in DMF and DMSO, the inclusion of Cs can be done by Spin or Deep coating [18, 101-103]. Once this is done, a heat treatment is carried out whose temperature will depend on the perovskite and the desired phase. LIU *et al.*, reported obtaining the perovskite CsP-bBr₃ by this technique, dissolving PbBr₂ in DMF and then adding this solution on an FTO substrate previously adhered in the spin coater, the coated FTO substrate was dried and dipped in a 30 mg solution of CsBr in methanol, varying the time from 5 to 15 minutes, for its subsequent heat treatment at 180 °C, generating smooth and dense films with excellent stability under atmospheric conditions [104]. The synthesis of CsP-bxSn_{1-x}I₃ has been reported using the two-step technique, applying a first layer of PbI₂ in DMF by spin-coating followed by immersion for 6 hours in a solution of CsI and SnI₂ in anhydrous methanol, followed by interation of the two-steps technique for making perovskite reached a stability in PSCs of up to 30 days [102]. Figure 6 illustrates the two-steps technique for making perovskite films.



Figure 6: Schematic procedure of the two-steps technique for obtaining perovskite films.

4.3 Vacuum processing

Vapor deposition is a technique used to obtain perovskite ABX₃ films that does not have the limitations of the solubility limits of solution techniques (one and two-step techniques), in addition to allowing more precise control of the thickness of the films generated, the homogeneity of perovskites, grain limits and their reproducibility [27, 105-107]. In 2017 *FROLOVA et al.*, reported obtaining a compact and homogeneous 300 nm layer by means of the co-evaporation technique using CsI and PbI₂ as precursors for obtaining CsPbI₃ with excellent photochemical and thermal properties [108]. LI et al., reported obtaining CsPbBr₃ / CsPb₂Br₅ perovskites by precisely controlling the film thickness of the precursor materials [109]. Another technique assisted by steam is the sequential vapor deposition, which consists of forming a first layer of CsX precursor, to later cover it with PbX₂, efficiently controlling the thickness of the resulting perovskite, this technique was used by TONG *et al.*, who with his workgroup obtained CsPbBr₃ perovskite with controlled grain sizes and passivation of grain boundaries, in addition to improving cargo carrier transport and achieving 45% controlled humidity stability for over 2000 hours) [28]. LUO *et al.*, obtained CsPbBr₃ by means of the assisted evaporation of Br on the perovskite of CsPbI₃, obtaining in a first stage the perovskite CsPbI₃ by means of the one-step technique, followed by the exchange of halogen by the assisted evaporation of Br [23]. Co-evaporation vacuum process (A) and vapor assisted deposition technique (B) for making perovskite films are

shown in Figure 7.



Figure 7: Co-evaporation vacuum process (A) and vapor assisted deposition technique (B) for making perovskite films.

5. INORGANIC PEROVSKITES

The inorganic perovskites that show the best efficiencies in PSCs, like hybrid perovskites, have the cation of Pb^{2+} as substituent B, while Cs^+ is the most used substituent A, in addition, they present the best results in terms of efficiency. On the other hand, the halides used are I, Cl or Br. Next, an analysis of the stability and efficiency of solar cells of the perovskites $CsPbI_3$, $CsPbBr_3$, $CsPbBr_2$, $CsPbBr_2I$, and $CsSnX_3$, which have better optical properties, as well as greater stability in the crystal structure, is presented.

5.1 Pure CsPbl₃, and CsPbBr₃ perovskites

The inorganic perovskite of CsPbI₃ is the one with the highest efficiency in PSCs so far, with a record that exceeds 18% efficiency. The high efficiency values achieved using this structure are due to the optical band gap of ~ 1.73 eV. The absorption that this perovskite presents has been reported between 700 and 750 nm in its active phase [75], ideal for photovoltaic applications, despite this, the cubic phase is not very stable at room temperature, once obtained at high temperatures it presents a rapid rearrangement of its structure by decreasing temperature, passing through three photoactive black phases, α -cubic, β - tetragonal, γ - orthorhombic and finally decaying to the inactive yellow phase δ -orthorhombic, the latter the most stable at room temperature, this it is explained by the low tolerance factor of Goldschmidt that the α phase presents, which is 0.807, just above the minimum limits necessary to formalize the perovskite [22, 26, 33, 71, 110].

The inorganic perovskite of CsPbBr₃, for its part, has proven to be a structure of greater stability than its CsPbI₃ counterpart, with a tolerance factor of 0.824. This perovskite has greater stability under environmental conditions, its active phase has a reddish coloration, so the absorption of this compound ranges between 520 and 550 nm in its cubic phase, in addition to presenting an optical band gap of 2.3 eV, which declines up to 4 eV, once it is completely degraded. The temperature required to obtain the cubic phase of this perovskite has been reported between 250 °C and 350 °C [28, 106, 111, 112].

Various works have focused on maintaining the cubic phase for as long as possible, investigating the

method of obtaining it, controlling the dimensions, the inclusion of various substituents in A and B as dopants, or the use of protective films (encapsulation) to delay atmospheric effects, or the use of carbon as a conductive electrode as a hydrophobic barrier [45, 113, 114].

5.2 CsPbl₃ perovskite

The use of HI or HPbI₃ as a precursor in the method of obtaining one and two steps, has been shown to improve the stability of the active phase of perovskite CsPbI₃ and allow it to be obtained at lower temperatures [25], XIANG *et al.*, reported obtaining CsPbI₃ using HI as a catalyst, which allowed the generation of stable perovskite for 2 months and 300 hours under irradiation, using carbon as the conductive electrode [98]. In the work reported by XU *et al.*, they use 4 (1H) -pyridinethione as an additive to promote crystallization through the interaction of S-Pb, which allows obtaining this α -cubic perovskite at low temperature (90-100 ° C) and keep it stable for 20 days, with a cell efficiency of 85%, compared to the first day of testing [115].

ZHANG *et al.*, reported obtaining CsPbI₃ with a stability of 1000 hours. Obtained by using phenyl-C61butyric acid methyl ester in mixture with chlorobenzene, as an antisolvent, it used methylammonium iodide as a mediator, since it allows to improve the quality of the films, and it is removed by heat treatment at 350 °C, generating PSCs with up to 16% efficiency [75]. Another additive used during the synthesis of this perovskite is phenylethylammonium iodide which hinders the transition to the inactive phase thanks to the steric effect of the additive once the crystals have formed [20]. Choline iodine was reported to stabilize perovskite and improve its electronic communication with the electron transport layer in the work presented by WANG *et al.*, who built a PSCs using this additive; It remained stable at lighting for 500 h, with only a loss of 5% of its efficiency [24]. The use of SCN⁻ as an additive to stabilize the perovskite α -CsPbI₃ was reported by YAO *et al.*, who compared to the perovskite obtained without the catalyst reported an increase in efficiency from 15.36 to 17.04%, as well as indicating that perovskite with additive took longer to progress to the yellow orthorhombic phase [85]. CHEN et al., obtaining highly efficient CsPbI₃ Quantum Dots (QD) based PSCs using mesoscopic TiO₂ as the ETM; they reported a fabricated device with the best PCE value reaching 14.32% (reverse scan) combined with an unprecedented high current density at short crcuit (J_{s-c}) of 17.77 mA cm⁻² [116].

5.3 CsPbBr₃ perovskite

In the work reported by ZHANG *et al.*, passivation of perovskite film defects was achieved using the ionic liquid 1-butyl-2, 3-dimethylimidazolium chloride, improving efficiency up to 61.3% compared to that obtained without the ionic additive, the perovskite obtained with the additive, maintained a stability under relative humidity of 70% at 20 ° C for 30 days with carbon as a conductive electrode [117]. Another additive used to promote the quality of perovskite crystals is the application of BiBr₃, prior to the application of the CsBr coating in the two-step technique, reported by PEI *et al.*, who obtained a stability of 90% PCE per 1000 h. using a carbon electrode [118].

WANG *et al.*, obtained the perovskite CsPbBr₃ by the two-step technique and using NH₄SCN as additive in the precursor solution of PbBr₂, followed by immersion in CsBr in methanol, generating a dense and homogeneous film with a stability of the cells greater than 200 hours under lighting [119]. BU *et al.*, reported obtaining this perovskite using the two-step technique, which was assembled in an HTL-free cell; the carbon used as an electrode was modified with polyaniline / graphite, which gave it a stability of up to 50 days at 80% relative humidity, with a 47% higher efficiency than unmodified carbon cells, this stability is attributed to the effect of the carbon modified as a better hydrophobic insulator [120].

XIANG *et al.*, prepared by vapor deposition the perovskite of CsPbBr₃ applying a first treatment of 320 ° C for 20 minutes, followed by a second treatment for 40 minutes at different temperatures, finding that a second treatment of 300 ° C achieves higher efficiency, coupled with excellent thermal stability of 40 days stored at 100 °C [27]. Table 1 presents a relationship of the architecture of different PSCs cells based on per-ovskites CsPbI₃ and CsPbBr₃, their stability and efficiency.

Architecture	Testing or storage	J _{sc}	Voc	FF	PCE	Reference
	time	(mA*cm²)	(V)	%	%	
CsPbI ₃						
FTO/TiO ₂ / CsPbI ₃ /Carbon	60 storage days	18.5	0.79	69.0	9.5	[98]
FTO/ TiO ₂ / CsPbI ₃ /PTAA/Au	60 storage days	18.95	1.059	75.1	15.07	[20]
FTO/PPTA/ CsPbI ₃ / PCBM layer/Ag	20 storage days	17.38	1.08	73.67	13.88	[115]
FTO/ TiO ₂ / CsPbI ₃ /Spiro-OMeTAD/Au	500 hours of testing	20.03	1.11	82	18.4	[24]
FTO/ TiO ₂ / CsPbI ₃ /PTAA/Au	Not reported	20.34	1.09	77	17.04	[8]
	1000 hours of tes-					
ITO/SnO ₂ / CsPbI ₃ / Spiro-OMeTAD/Au	ting	20.1	1.06	75.3	16.04	[75]
				Not re-		
FTO/ TiO ₂ /CsPbI ₃ /Spiro-OMeTAD/Au	100 hours of testing	17.77	0.84	ported	14.32	[116]
CsPbBr ₃						
FTO/ TiO ₂ /CsPBr ₃ / Mixed-carbon	30 storage days	7.16	1.357	72.97	7.09	[113]
FTO/ TiO ₂ /CsPbBr ₃ /Spiro-						
OMeTAD/Au	60 storage days	7.71	1.37	81	8.65	[106]
FTO/ TiO ₂ /CsPbBr ₃ /Carbon	30 storage days	7.45	1.61	83	9.92	[117]
	1000 hours of test-					
FTO/ TiO ₂ /CsPbBr ₃ /Carbon	ing	7.84	1.39	80	8.73	[118]
ITO/ TiO ₂ /CsPbBr ₃ /Spiro-						
OMeTAD/Au	200 hours of testing	7.76	1.375	79.31	8.47	[119]
FTO/ TiO ₂ /CsPbBr ₃ /Carbon	50 storage days	6.87	1.59	81.21	8.87	[120]
FTO/ TiO ₂ /CsPbBr ₃ /Carbon	40 storage days	7.37	1.545	82.2	9.35	[27]

Table 1: Pe	erformance and	stability time	of PSCs with	CsPbI ₂ and	d CsPbBr ₂	perovskites.
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J_{SC} = Current density at short circuit; V_{OC} = Voltage at open circuit; FF = Fill factor; PCE = Power-conversion efficiency

5.4 Halide mixture in CsPb perovskite

The band gap of 1.73 eV of its own and CsPbI₃ is ideal for use as an adsorbent layer in PSCs, however its low thermal stability limits its long-term application in these devices [75]. On the other hand, the perovskite of CsPbBr₃ is more stable under normal atmospheric conditions it presents a band gap of 2.3 eV [118], so the combination in different proportions of the halides I and Br in the structures allows to combine the intrinsic properties of these semiconductors, and allow to substantially improve the stability thus as a greater absorption of the electromagnetic spectrum. The CsPbIBr₂ perovskite has an absorption around 600 nm with 2.05 eV optical band gap, as well as a stability superior to that of the ABI₃ perovskites and a better efficiency than the ABBr₃ perovskites, which exceed 12% [103,121]. For its part, the perovskite composed with CsPbI₂Br further reduces its energy gap by around 1.9 eV and it improves its thermal stability, although to a lesser extent than its CsPbIBr₂ counterpart with an absorbance close to 650 nm and efficiencies greater than 16% [19, 57, 122]. For these reasons year after year more work is done focused on improving both the properties of these absorbent layers and their performance in PSCs. It should be clarified that both structures can be obtained by following the same procedures as singles halide perovskites.

In an effort to increase the stability of the absorbing layer, NiO_x and CeO_x charge transport films were used to isolate the perovskite CsPbIBr₂ from the effects of humidity and temperature according to what was reported by YANG et al.; they reported that this dispositive achieved stability for 500 hours (stored at 45-50% of humidity) [123]. ZHU et al., obtained a perovskite of CsPbBr₃ using the one-step technique followed by the addition of CsI in methanol to obtain CsPbIBr₂ methodology that allowed obtaining a film that presented few limits grain, high crystallinity and that in cell allowed to maintain 90% of the conversion efficiency for 60 days in environmental conditions of humidity and temperature [124]. ZHANG et al., synthesized the CsPbIBr₂ perovskite using the one-step technique, using diethyl ether as an anti-solvent and a guanidinium additive to passivate the structure defects, obtaining a perovskite with a stability of 100 hours and an efficiency of 9.17%, which is high for this structure [94]. As already mentioned, the TiO_2 antasa phase is the semiconductor that has been most used as ETL in PSCs, it is used as a TiO_2 compact film (c-TiO₂) or as a TiO₂ mesoporous film (m-TiO₂), a photoactive inorganic perovskite of CsPbI₂Br on TiO₂ films was deposited

using a gradiente thermal annealing method (GTA) [125, 126] and PCE values of 14.5% and 16.07% were obtained, while PCE values of 15.25% and 15.69% were obtained when doping the CsPbI₂Br perovskite with In³⁺ and Eu²⁺cations [127, 128]. Some researchers report that precalating the glass-FTO/c-TiO₂ substrate at 90 °C and using tammonium hexafluorophosphate (NH₄PF₆) as precursor additive to modify the grain size of the CsPbI₂Br perovskite it is possible to obtain a photovaitaic device with an efficiency of 10.1% [129]. On the other hand, tin oxide (SnO₂) has been used as ETL in solar cells containing CsPbI₂Br perovskite and using ethyl acetate as anti-solvent or a programmable crystallization method to obtaining PCE values from 14.33 % to 16.58% andy J_{sc} values from 13.61 to 16.23 mAcm⁻² [130-132]. LIU *et al.*, obtained the inorganic perovskite of CsPbIBr₂ by means of the two-step technique assisted by vapor deposition, using copper (II) phthalocyanine (CuPC) and carbon as an electrode transporting film, which provides better stability to both perovskite and the device in general resisting for one month at 60 °C [107]. Table 2 collects results from works reported for PSCs in which the stability as a function of perovskite storage or test time of CsPbIBr₂ and CsPbI₂Br can be seen.

5	2	2				
Architecture	Testing or storage	J_{sc}	V _{oc}	FF	PCE	Refe-
	time	(mA*cm²)	(V)	%	%	rence
FTO/NiO _x /CsPbIBr ₂ /ZnO@C ₆₀ /Ag	10 hours of testing	15.1	1.1	75.5	12.6	[133]
FTO/TiO ₂ / CsPbIBr ₂ /Spiro-OMeTAD/Au	100 hours of testing	10.24	1.2	74.6	9.17	[94]
FTO/TiO ₂ /CsPbIBr ₂ /CuPc/Carbon	720 hours of testing	9.65	1.236	63.1	8.76	[107]
ITO/NiO _x /CsPbIBr ₂ /CeO _x /Ag	500 hours of testing	8.76	1.01	63.35	5.60	[123]
FTO/TiO ₂ /CsPbIBr ₂ /Carbon	60 days of storage	10.71	1.169	66	9.16	[124]
FTO/TiO ₂ /CsPbI ₂ Br/PCBM/Ag	500 hours of testing	15.33	1.22	78.8	14.78	[134]
FTO/c-TiO ₂ /m-TiO ₂ /CsPbI ₂ Br/ Spiro-						
OMeTAD /Ag	3 hours of testing	15.8	1.27	78.0	15.5	[125]
ITO/c-TiO ₂ /CsPbI ₂ Br/Spiro-OMeTAD /Au	120 hours of testing	16.79	1.23	77.81	16.07	[126]
FTO/c-TiO2/m-TiO2/Doped-CsPbI2Br/	>1600 hours of					
CuSCN/rGO/Au	testing	15.91	1.28	74.85	15.27	[127]
	30 storage days in					
FTO/TiO ₂ /CsPbI ₂ Br/Spiro-OMeTAD/Au	air	15.44	1.25	79.00	15.25	[128]
	25 storage days in					
ITO/SnO ₂ /CsPbI ₂ Br/Spiro-OMeTAD/Ag	N ₂ atmosphere	16.23	1.21	76.29	15.07	[130]
ITO/SnO ₂ :C ₆₀ -EDA/CsPbI ₂ Br/Spiro-	65 storage days in					
OMeTAD/MoO/Ag	dry air	15.41	1.34	80.30	16.58	[131]
ITO/ ZnO:Cs ₂ CO ₃ / CsPbI ₂ Br/PCBM/Ag	200 hours of testing	16.34	1.28	78.42	16.42	[97]
FTO/NiO _x /CsPbIBr ₂ /ZnO@C ₆₀ /Ag	300 hours of testing	15.2	1.14	77	13.3%	[121]
ITO/SnO ₂ -ZnO/ CsPbIBr ₂ /Spiro-OMeTAD						
-MoO ₃ /Ag	360 hours of testing	15	1.23	78.8	14.16	[122]
ITO/SnO _x / CsPbIBr ₂ / Poly (DTSTPD-r-	900 hours of inter-					
BThTPD)Au	mittent testing	14.25	1.4	77	15.53	[135]
	1000 hours of test-					
FTO/TiO ₂ / CsPbIBr ₂ / Spiro-OMeTAD/Au	ing	15.32	1.32	83.29	16.79	[93]
	2.5 hours of testing					
ITO/ZnO/ CsPbIBr ₂ / Spiro-OMeTAD/Ag	at 200 °C	14.9	1.22	75.3	13.7	[87]
PDOT:PSS/Al:ZnO/CsPbIBr2/tBCA/PTA						
A/MoO ₃ /Ag	60 storage days	15.87	1.26	75.41	15.08	[136]

Table 2: Performance and	stability time	of PSCs with	CsPbI ₂ Br and	CsPbIBr ₂
	2		4	

 $J_{SC} = Current \ density \ at \ short \ circuit; \ V_{OC} = Voltage \ at \ open \ circuit; \ FF = Fill \ factor; \ PCE = Power-conversion \ efficiency; \ CuSCN = Copper \ thiocyanate; \ C_{60}-EDA = Fullerene-ethylenediamine; \ rGO = Reduced \ graphene \ oxide$

The synthesis of CsPbI₂Br by the one-step technique using DMSO ducts, which allows the formation of

large-grain films with low defect density, and a longer life time for charge carriers, as well as better stability of up to 500 h according with GUANNAN *et al.* [135]. The use of CaCl₂ as an additive in the formation of the CsPbI₂Br perovskite by the one-step technique allows obtaining more stable structures with a prohibited band of 1.91 eV as well as good performance in PSCs, reaching a competitive efficiency of 16.79%, which is maintained at 90% after 1000 hours of continuous testing [93]. Processing at low temperatures is feasible to obtain perovskite as demonstrated by Xia Yang and his team, who obtained the absorbent layer by the onestep method at 120 ° C. This was used to assemble a flexible cell which reached an efficiency 15% maximum that declined 7, 9 and 86% after 60 days stored under environmental conditions, 360 hours at 85 ° C and 30 h at 65% humidity, respectively. The cell structure was formed using tert-butyl cyanoacetate (T-BCA), PTTA and MoO₃ as gap carriers, ZnO₂ aluminum doped and PEDOT: PSS as gap carriers [136].

Thermal stabilization and optimization of CsPbI₂Br-based PCSs have also been tested by applying ZnO₂ doped with Cs₂CO₃ as an electron-bearing film that allows a good extraction of these charge carriers, resulting in an increase in efficiency, coupled with the isolation of perovskite that increases its stability ZnO: Cs₂CO₃ maintains efficiency at 93% for 200h. vs 73% who maintained the device that used only ZnO₂ [97]. Another film used to improve electron transport is ZnO₂ @ C₆₀ which in 2018 achieved a photoconversion efficiency of 13.3% with a stability that reached 360 hours with only 20% loss of initial efficiency [59], results comparable to those presented by YAN *et al.*, who reported a stability of 300 hours with a loss of 15% of the photoconversion efficiency (14.16%) using SnO₂-ZnO as an electron carrier [122]. For his part, GOU *et al.*, reported the device based in CsPbI2Br, the poly [(dithiene [3,2-b: 2 ', 3'-d] silolethiene [3,4-c] pyrrole-4,6-dione) -random- (2,2'- bithiophenethiene [3,4-c] pyrrole-4,6-dione) (poly (DTSTPD-r-BThTPD)) (and as an electron acceptor SnCl₂ device that reached a PCE of 15.53% and remained under intermittent illumination for 900 h, maintaining 71% of the initial efficiency [135]. In general, a greater number of works focused on CsPbI₂Br are reported due to their lower bandgap. that allows obtaining greater efficiencies in PSCs although they have less stability, on the other hand, CsPbIBr₂ have better stability but less efficiency, so much of the effort is focused on increasing efficiency.

5.5 CsSnX₃ perovskite

In the search to obtain lead-free perovskites, the use of Sn^{2+} has been tried to substitute it, since said element presents serious damage to the environment, thus the CsSnI₃, CsSnBr₃ and CsSnCl₃ perovskites are presented as a greener option than their counterparts. These include lead, these structures have an optical band interval along the near and visible infrared region, with absorbance at 750, 610 and 420, respectively. However, Sn^{2+} has short periods of functionality as a photoactive layer due to a rapid oxidation of Sn^{2+} to Sn^{4+} . In the case of the perovskite $CsSnI_3$, it has a band gap of 1.3 eV, which is ideal for its application in photovoltaic cells [40, 89, 137-139]. In this sense, ZHANG et al., stabilized the $CsSnI_3$ perovskite in a reducing environment by using cobaltocene, which is a large electron donor, which prevents Sn²⁺ oxidation, which results in increased stability of CsSnI₃, however, it presents a low efficiency of 3%, however this is an encouraging result to replace lead in these devices [137]. The highest efficiency reported so far for $CsSnI_3$ is 5.03% obtained by WANG et al. To do this, they synthesized perovskite in a single step using triphenyl phosphite as an antioxidant additive to stabilize perovskite, which allowed its analysis under illumination for 400 minutes, which is considerable for this type of perovskites [140]. Another work on the development of cells based on perovskite $CsSnI_3$ is that reported by ZHU *et al.*, who obtained the perovskite $CsSnI_3$ using the evaporative assisted solution method. The photovoltaic device had a maximum yield of 2.23% and an average yield of 1.93% that presented a decay at 70% of its initial efficiency at 70% humidity, after 60 min [141]. LEI et al., synthesized the perovskite $CsPb_xSn_{1-x}I_3$, with a band gap between 1.3 and 1.7 eV observing a better electrical conductivity when increasing the Sn^{2+} concentration; although they did not build cells, their research provides an applicability option to these devices with a lower concentration of lead [110, 142].

The perovskite $CsSnBr_3$ presents a band gap value around 1.9 eV, a thermal stability higher than 437 °C and a transition to its inactive phase that takes 48 hours at room temperature and around 60% humidity [143]. However, its preparation using additives such as SnF_2 has allowed $CsSnBr_3$ to be obtained with an optical band gap of 1.73 eV by reducing the gap between the valence band and the fermi level, coupled with limiting the formation of Sn^{2+} to Sn^{4+} [145]. SABBA *et el.*, reported PSCs of this structure prepared without SnF_2 and with this additive, demonstrating that the efficiency increases slightly from 0.1 to 0.91%, in turn they observed that if Br^- is included in the perovskite of $CsSnI_3$ the efficiency improves reporting a maximum efficiency of 1.67% for the perovskite $CsSnIBr_2$, the latter with a prohibited band of 1.75 eV [144]. ISLAM *et al.*, synthesized the perovskite $CsSnCl_3$ in order to improve its optoelectronic properties, since this structure has a band gap of 2.8 eV, which rules it out of applications in photovoltaic cells, so by including metals such as Cr or Mn sought to increase absorption in the visible spectrum, demonstrating that the valence band of Mn-

doped $CsSnCl_3$ samples shifts slightly towards the region of highest energy, which facilitates the promotion of electrons towards the conduction band, showing better photoconductivity and greater absorption than pure perovskite from $CsSnCl_3$ [86].

Architecture	Testing or storage	J _{sc}	V _{oc}	FF	PCE	Reference
	time	(mA*cm²)	(V)	%	%	
FTO/TiO ₂ /Al ₂ O ₃ / CsSnI ₃ /NiO/Carbon	100 hours of stor- age	18.24	0.36	46	3.0	[137]
FTO/TiO _{2/} CsSnIBr ₂ /Al ₂ O ₃ /CsSnI ₃ /Carbon	9 hours of testing	17.4	0.31	57	3.2	[143]
ITO/PEDOT:PSS/CsSnI ₃ /PCBM/Ag	30 days of storage under N_2	23.792	0.42	No report- ed	4.13	[140]
ITO/ TiO ₂ / CsSnI ₃ / Spiro- OMeTAD:TBP/Au	No reported	27.67	0.201	29	1.66	[144]
ITO/ TiO ₂ / CsSnI ₂ Br/Spiro- OMeTAD:TBP/Au	No reported	15.06	0.289	38	1.67	[144]
ITO/ TiO ₂ / CsSnIBr ₂ / Spiro- OMeTAD:TBP/Au	No reported	11.57	0.311	43	1.56	[144]
ITO/ TiO ₂ / CsSnI ₃ / Spiro-OMeTAD: TBP/Au	No reported	3.99	0.410	58	0.95	[144]

 J_{SC} = Current density at short circuit; V_{OC} = Voltage at open circuit; FF = Fill factor; PCE = Power-conversion efficiency

Great efforts have been focused on optimizing the fabrication processes of solar cells with different structures based on ABX₃ inorganic perovskites, using different methods like spin coating and vacuum deposition, controlling heat treatment, grain size of the photoactive phase, and the use of both organic and inorganic additives with the aim to get photovoltaic diapositives with high efficiency (PCS), filled factor (FF), open circuit voltage (Voc), and short circuit current density (Jsc) with high stability under environmental conditions of temperature and moisture. Table 4 show a summary of the best solar cells based on inorganic perovskite obtained until now; the three inorganic halide perovskites solar cells that present the best values of efficiency at high testing time are CsPbI₃, CsPbIBr₂, and CsPb₂Br, with the architectures of FTO/TiO₂/CsPbI₃/Spiro-OMeTAD/Au, FTO/TiO₂/CsPbIBr₂/Spiro-OMeTAD/Au, and FTO/c-TiO₂/m-TiO₂/Doped-CsPbI₂Br/ CuSCN/rGO/Au, respectively.

Architecture	Testing or storage	J _{sc}	Voc	FF	PCE	Referen-
	time	(mA*cm ²)	(V)	%	%	се
FTO/ TiO ₂ / CsPbI ₃ /Spiro-OMeTAD/Au	500 hours of testing	20.03	1.11	82	18.4	[24]
FTO/ TiO ₂ / CsPbI ₃ /PTAA/Au	Not reported	20.34	1.09	77	17.04	[8]
FTO/TiO ₂ / CsPbIBr ₂ / Spiro-OMeTAD/Au	1000 hours of testing	15.32	1.32	83.29	16.79	[93]
ITO/SnO2:C60-EDA/CsPbI2Br/Spiro-	65 storage days in dry	15 41	1.24	00.20	16.59	[121]
OMeTAD/MoO/Ag	air	15.41	1.34	80.30	10.58	[131]
ITO/ ZnO:Cs2CO3/ CsPbI2Br/PCBM/Ag	200 hours of testing	16.34	1.28	78.42	16.42	[97]
ITO/c-TiO ₂ /CsPbI ₂ Br/ Spiro-OMeTAD /Au	120 hours of testing	16.79	1.23	77.81	16.07	[126]
ITO/SnO ₂ / CsPbI ₃ / Spiro-OMeTAD/Au	1000 hours of testing	20.1	1.06	75.3	16.04	[75]
FTO/c-TiO ₂ /m-TiO ₂ /Doped-CsPbI ₂ Br/	> 1600 hours of test-	15.91	1.28	74.85	15.27	[127]
CuSCN/rGO/Au	ing					

Table 4: PSCs made with ABX₃ inorganic halide perovskites with the higher efficiency and testing time.

6. CONCLUSIONS

Inorganic perovskites are a promising alternative to supply the most widely used organometallic perovskites for the development of solar cells, so the focus of this review was to know the factors that affect these structures, the methods of obtaining them, the architecture with which photovoltaic devices, their components and processes of generation and load mobility are manufactured, as well as trends to improve both stability and efficiency. In this work we compile the photoconversion efficiencies reported in different investigation reports, as well as the stability that these devices present under light irradiation or due to their storage time, under different conditions of temperature and humidity. Various studies carried out to adjust the perovskite band gap to improve its capacity as an adsorbent film, by doping or substitution of metal cations A^+ and B^{2+} , or by mixing halides I and Br, were analyzed. The techniques used to stabilize the photoactive phases of perovskites, which range from the method of obtaining, the use of additives, heat treatments, the composition of perovskites, the application of various materials as charge carriers, mainly inorganic, which manage to stabilize the active phase. On the other hand, the use of carbon as an electrode generates a hydrophobic layer that stabilizes the PSCs and time of use and storage is increased. The total substitution of Pb in the perovskite structures is also addressed since, due to the contamination that this element causes to the environment. In general, inorganic perovskites can considerably improve their performance using all these considerations, being the halides mixture the most common route promising to achieve an efficient and stable structure, so a large number of papers show increasing interest in the use of $CsPbI_{x-y}Br_y$ perovskites, and cell components to optimize these devices.

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8. CONFLICTS OF INTEREST

The authors have no conflicts of interest to declare.

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