# **Additive Engineering**

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Additive engineering based on quantum dots containing different functional groups introduced in the precursor. This allows the functional group to chelate under coordinated Pb vacancy or iodine defect sites at the surface of the perovskite layer.

Keywords: additive ; MAPbI ; stability ; ambient ; inert ; perovskite solar cell ; efficiency

# 1. Introduction

Alternative renewable energy sources are considered reliable options for long-term usage due to the limited availability of traditional energy resources (i.e., coal, oil, and gas). Since solar energy is abundant and free (i.e., it comes from a natural source), it is referred to as clean and green energy. Also, solar energy can help reduce air pollution and global warming caused by greenhouse gas emissions if low-cost manufacturing is achieved <sup>[1]</sup>. This notion of utilizing solar energy further led to the development of photovoltaic technology, resulting in three generations. The first generation of photovoltaic technology was silicon wafer-based solar cells. However, they were not commercially successful due to their high cost. In contrast, second generation solar cells offered a low cost, but compromised in efficiency. Hence, the third generation was developed to be cost-effective and efficient. In this context, perovskite solar cells are considered advanced level third generation photovoltaic technology that could offer low-cost processing with ease of manufacturing and equally high efficiency when compared to their traditional counterparts <sup>[2]</sup>. This is why the research community has paid attention to perovskite solar cells in the last decade.

"Perovskite" is the name of the mineral CaTiO<sub>3</sub> discovered in the Ural Mountains of Russia; it is named after the Russian nobleman and mineralogist, Lev Perovski. Nevertheless, this term is being used for all compounds, with the general formula ABX<sub>3</sub>, with the same crystal structure as CaTiO<sub>3</sub> or derived from this structure. These materials consist of two cations. Cation A is 12-fold coordinated by anion X, and cation B is 6-fold, where X can be either oxygen or a halide. In terms of perovskite solar cells, the most commonly used A-site cations are MA/[CH<sub>3</sub>NH<sub>3</sub>]<sup>+</sup>, FA/[H<sub>2</sub>NCHNH<sub>2</sub>]<sup>+</sup>, Rb<sup>+</sup>, Cs<sup>+</sup>, B site cations are Pb<sup>2+</sup>, Sn<sup>2+</sup>, Ge<sup>2+</sup> While the X site is halogen atoms, I<sup>-</sup>, Cl<sup>-</sup>, Br<sup>-</sup> [3]. Among many different perovskite absorber layers, methyl ammonium lead iodide (MAPbl<sub>3</sub>) is the most prominently studied hybrid organic-inorganic perovskite for perovskite solar cell application. MAPbl<sub>3</sub> offers several unique properties and advantages to be applicable in perovskite solar cells (PSC). It has a suitable bandgap~1.6 eV, a high absorption coefficient (the absorption coefficient of MAPbl<sub>3</sub> lies in the range of  $10^4$ – $10^5$  cm<sup>-1</sup>, which is more than one order of magnitude larger than that of silicon for the visible light spectrum). The high absorption coefficient of MAPbl<sub>3</sub> allows it to absorb in the low light region. Moreover, a high absorption coefficient allows light to be absorbed by a thin film (generally in the range of 0.3-0.6 µm) of the perovskite layer, while crystalline silicon-based solar cells are usually made thicker ~300 µm. This, in turn, reduces the quantity of required material, thereby reducing the cost. Besides, MAPbl<sub>3</sub> can be easily solution-processed to produce efficient solar cells, which further lowers manufacturing costs. Moreover, MAPbl3 can result in high open-circuit voltages  $(V_{oc})$ , low  $V_{oc}$  loss/deficit, and low exciton binding energy: 2–70 meV, resulting in better charge transport with decent charge carrier motilities of 2–66 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>, long diffusion lengths of charge carriers  $\sim 1$  um. Because of these properties, scientists and researchers have recommended the fabrication of perovskite solar cells using MAPbl<sub>3</sub> as the active layer <sup>[4]</sup>  $[\underline{S}][\underline{G}]$ . Nevertheless, long-term operational stability has been a significant factor that acts as a blockade for the commercialization of PSCs. In comparison, the traditionally made silicon-based solar cells remain operational for up to 20-25 years. On the contrary, perovskite solar cells are stable for a few hundred hours to a maximum of one year [4]. Thus, PSCs face serious stability issues compared to already available PV technology in the market. These stability issues involve both extrinsic and intrinsic challenges to overcome.

# 2. Organic Additives

Organic additives are the most common types of additives used in perovskite solar cells. These organic additives can be further categorized based on N donor, O donor, and S donor atoms. These electron donor atoms can bind/coordinate with the  $Pb^{2+}$  species, resulting in adduct formation, passivating the grain boundaries and, thus, improving efficiency and stability. Subsequently, based on the electron-donating atoms, additives can be further subcategorized as amines, nitriles belonging to the N donor atom, amides, acids, acetates, alcohol, esters, and ethers, belonging to oxygen donor atoms and sulfides, thiocyanates belonging to the S donor atom [I].

Following the notion of amine-based additives, the non-stoichiometric use of methylammonium iodide as an amine additive was advised in the early phases of developing perovskite solar cells. It was established that adding excess MAI reduces trap density, increases PL lifetime, and further increases PCE regardless of fabrication methods [8][9][10]. Likewise. some other organic amine derivatives, such as benzylammonium iodide (BAI) and phenethylammonium iodide (PEAI), have also been investigated as additives and were found to improve light harvesting properties and exciton lifetime with reduced charge recombination [11][12]. Moreover, it was discovered that, if hydrophobic cations containing amine additives are employed, such as hexylamine hydrochloride (1-HH), 1,6-diaminohexane dihydrochloride (1,6-DD) and phenylhydrazinium iodide (PHAI), it may even work as a protection for the MAPbl<sub>3</sub> film against moisture, improving the stability in ambient conditions <sup>[13][14]</sup>. Furthermore, nitrogen-containing heterocyclic amines play a great role in improving stability <sup>[15]</sup>. For instance, the introduction of 4,4'-bipyridine in MAPbI<sub>3</sub> demonstrated complex formation with PbI<sub>2</sub>, thus improving intrinsic stability against illumination [16]. Additionally, it has been shown that the presence of N atoms prevents the loss of volatile species from the perovskite film and acts in a similar way as polymeric passivation coating. Further, such a passivation effect was visible, morphologically (in the SEM cross-section images and energy dispersive X-ray analysis), when PVC was added in MAPbl<sub>3</sub> [12]. Later, scientists designed pyridine derivatives with units containing different multifunctional groups. One such additive is (C<sub>60</sub>-PyP), which contains C<sub>60</sub> units that are hydrophobic in nature and pyridine units, which chelates  $Pb^{2+}$  by donating the lone electron pair on the N atom. It is known that uncoordinated Pb<sup>2+</sup> ions are considered trap states at grain boundaries and can block charge extraction [18]. Thus, grain boundaries are passivated by assisting coordination interactions with the Pb<sup>2+</sup> ion of MAPbl<sub>3</sub> and PyP unit that further improved short circuit current density  $J_{sc}$  and eliminated ion migration. While the hydrophobic  $C_{60}$  unit does not let moisture directly affect the film [18]. Similar results were noticeable when pyridine-2-carboxylic lead salt (PbPyA<sub>2</sub>) and polyvinylpyrrolidone (PVP) additives were employed in MAPbI<sub>3</sub> precursor <sup>[19][20]</sup>.

Similar to N donor, O donor and S donor can also act as Lewis bases due to lone pair of electrons available to co-ordinate with Pb defect site. Additives containing O atom as the Lewis base may consist of various functional groups, such as carbonyls, carboxyl, carboxylates, ester, ethers, and alcohols <sup>[1]</sup>.

### 3. Inorganic Additives

Inorganic salts can be further explained as per the category in the periodic table, i.e., based on alkali metals, alkaline earth metals, transition metals, other metals, and other non-metals. These additives may exist in the form of inorganic halide salts or inorganic acids <sup>[21]</sup>.

Hydro halides are at the top, as hydrogen is the first element in group one of the periodic table. Soe et al. studied the incorporation of hydrohalic acids (HX, X=I, Br, CI) in MAPbI 3 precursor prepared using DMF. It was found that HX incorporation in the perovskite layer alters the bandgap and unit cell parameters. The HI addition compressed bandgap, whereas HBr widens the bandgap at high concentrations (20-25 vol%). Further, these changes were found correlated with the types of defects present in polycrystalline perovskite thin films combined with the structural strain induced in very small crystallites. Thus, it was concluded that these acids could influence crystallization rate, surface coverage, and improve morphology. However, HCl incorporation showed no influence on bandgap <sup>[22][23]</sup>. The same was verified by Yan and co-workers who investigated HI (hydroiodic acid) as an iodine quencher in MAPbI 3 precursor solution (prepared with isopropyl alcohol (IPA)), that further lead to chemically passivate grain boundaries and increase in PCE from 16.13% (pristine) to 18.21% (with HI additive) <sup>[24]</sup>. Likewise, alkali metals, such as Li, Na, and K, combined with halogen counterparts, have been inspected as additives for the MAPbI 3 light absorber layer. Grätzel and Friend co-workers demonstrated the influence of Nal (sodium iodide) on the optical, excitonic, and electrical properties of CH 3NH 3Pbl 3 perovskite prepared by a two-step sequential deposition technique. It was revealed that the Nal additive helps in the complete conversion of PbI 2 into CH 3NH 3PbI 3, leaving no unreacted PbI 2 and enhancing the crystallinity. Furthermore, KPFM measurements showed a reduction in work function (towards Au metal) for contact-potential difference (CPD) and proved the chemical passivation of perovskite surface due to additive incorporation in the precursor. Additionally, the hypothesis of partial substitution of Pb 2+ with monovalent cations Na was discarded, because replacing

Pb 2+ with the Na cations required high energy, which could not be obtained at room temperature, since additives and films were processed at room temperature <sup>[25]</sup>. Similar reports were published from Chu and co-workers who investigated alkali metal chloride additives (LiCl, NaCl, and KCl) in MAPbI 3 and suggested that mixed halide formation does not occur as Cl atoms easily evaporate due to the annealing process, improving the crystallinity and carrier charge transport <sup>[26]</sup>. However, as the size of the nuclei increases, the perovskite shows partial replacement and forms a perovskite with a double cation (i.e., in the case of RbX and CsX, where X represents halide) <sup>[27]</sup>. The partial substitution of cation was also observed when alkaline earth metals were employed as additives in the precursor. Hence, they are not the center of attraction here and it is suggested that alkaline earth metals are best suited to replace Pb due to their divalent nature <sup>[28]</sup>.

However, the most widely reported metal additive is PbI 2. Several groups have suggested using excess PbI 2 as an additive and studied the effect of the stoichiometric and non-stoichiometric composition of PbI 2 in MAPbI 3 perovskite <sup>[30]</sup> <sup>[31]</sup>. Reports suggest that the addition of excess PbI 2 in perovskite increases power conversion efficiency <sup>[30][32]</sup>. However, the influence of excess PbI 2 on photochemical stability is debatable <sup>[33]</sup>. Some research groups claim that excess PbI 2 passivates grain boundary <sup>[34]</sup>; thus, creates extra passivation <sup>[35]</sup>; enhancing performance and stability overall <sup>[31][36]</sup>. On the contrary, some reports showed that excess PbI 2 could increase PCE, but decreased PSC stability <sup>[37][38]</sup>. To resolve the issue, Stevenson et al. demonstrated that the stability associated with excess PbI 2 is affected by solvent coordination capacity with Pb atom, and suggested that excess PbI 2 can work as a stabilizer to improve intrinsic stability if the correct solvent (NMP) is chosen over commonly-used solvent DMF <sup>[39]</sup>. Other than PbI 2, adding PbCI 2 also resulted in an equally good perovskite film quality with improved performance, but the outcome was reported as a mixed halide <sup>[40][41]</sup>. Hence, it is not focused on here. Interestingly, Ngo and the group demonstrated a different approach by using PbS quantum dots as capping ligands in the precursor solution to control nucleation and morphology. This approach allowed improving crystallinity and enhanced grain size, resulting in improved solar cell performance <sup>[42]</sup>.

### 4. Outlook

Further developments are required while adopting the additive approach in the fabrication of MAPbI 3-based PSCs. So far, additives employed in the MAPbI 3 light-absorbing layer do not have any rule of selection. The majority of additives have opted from existing literature available for DSSCs or organic solar cells, utilizing the N/O/S donor atoms to combine with under-coordinated Pb 2+ sites to improve stability because of the Lewis acid–base reaction. Hence, the first and the topmost prospect is to design an additive that can help sustain MAPbI 3 under operational conditions, such as illumination, heat, moisture, oxygen, water.

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