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Addressing the Stability Challenge of Perovskite Solar Cells: The Potential of Ionic Liquid Incorporation for Improved Device Durability

Artur F. M. Farinha, Luís M. N. B. F. Santos, José C. S. Costa

CIQUP, Institute of Molecular Sciences (IMS), Department of Chemistry and Biochemistry, Faculty of Science, University of Porto, Rua do Campo Alegre s/n, P4169-007 Porto, Portugal (up201506214@fc.up.pt) ORCID 0000-0001-9857-3584 (lbsantos@fc.up.pt) ORCID 0000-0003-3040-0358 (jose.costa@fc.up.pt) ORCID 0000-0002-7134-8675.

Abstract

Perovskite solar cells (PSCs) have emerged as a promising technology for renewable energy generation due to their low cost and low carbon footprint compared to traditional silicon-based solar cells. However, some main challenges associated with PSCs lie ahead, namely their toxicity and lack of stability, particularly under factors such as light, temperature, oxygen, and humidity. This review focuses on the lack of stability of PSCs and the various ways it can be mitigated. We explore different methodologies, solution and vapor based, and different strategies for PSC production and enhancing. Furthermore, the potential of ionic liquids (ILs) as promising materials for improving the stability and performance of PSCs is highlighted. ILs have advantageous physicochemical properties that make them suitable as an additive or interfacial layer in PSCs. They optimize the interface contact, improve energy level matching, suppress ion migration, and increase hydrophobicity, which inhibits the decomposition of the device in humid environments. ILs have also been used as precursors in the solution-based fabrication of perovskite thin films for PSC applications, assisting in the perovskite crystallization. Several studies have shown that the incorporation of ILs in PSCs can increase stability, lifetime, and efficiency. The existing research indicates that ILs hold great promise as materials for improving the stability and performance of PSCs, which could have significant implications for the development of low-cost, renewable energy technologies.

Author Keywords: Perovskite Solar Cells, Ionic Liquids, Thin Films, Interface Engineering

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The significance of solar energy stems from its ability to provide a virtually limitless and clean source of renewable energy that can reduce dependence on finite fossil fuels, mitigate climate change, and promote sustainable development. One of the most efficient and practical ways to use the sunlight as an energy source is to convert it to electricity using solar cells. The most popular and efficient solar cells to this day are those made from thin wafers of crystalline silicon, which are also the oldest solar cell technology. However, a number of thin-film technologies, often described as emerging photovoltaics are in the research or development phase (Nayak et al. 2019; Ranabhat et al. 2016).

Perovskite Solar Cells

Perovskites are a group of materials whose crystalline structure follows a general chemical formula ABX_3. They exhibit a vast array of properties and applications. They can function as electrical insulators, conductors or semiconductors, and they have been found to exhibit properties such as piezoelectricity, giant and colossal magnetoresistance, and even superconductivity (Alonso et al. 2003; Ge et al. 2016; Hao et al., 2019; Park, Ha, and Lee, 2020; Rubel et al., 2016; Stoumpos et al., 2013; Su et al. 2021; Yadav and Elizabeth, 2015). The sheer diversity of these properties makes perovskites an incredibly valuable area of study, and they are widely used in many practical applications. They have emerged as a crucial component in many high-performance solid-state devices, such as capacitors, actuators, transducers, and optical modulators (Hao et al. 2019; Kovalenko, Protesescu and Bodnarchuk, 2017; Y. Zhang et al. 2019). Additionally, they play a vital role in catalysis (Hwang et al. 2017; Xu, Zhong and Shao 2019).

In 2009, the first perovskite-based photovoltaic device was published by Miyasaka's group. (Kojima et al. 2009). Since then, hybrid organic/inorganic perovskites, which are composed of a monovalent organic cation on position A, a divalent metal on B, and a halide anion on X, have been applied in perovskite solar cells (PSCs) (Burschka et al. 2013; Liu, Johnston and Snaith 2013; N. G. Park 2020; Saliba, Matsui, Domanski et al. 2016; Saliba, Matsui, Seo, et al. 2016), which are low-cost and have a low carbon footprint compared to traditional silicon-based solar cells. Despite their relative novelty, PSCs have already achieved exceptional power conversion efficiencies (PCE > 25%) (Burschka et al. 2013; Emami, Andrade and Mendes 2015; Liu, Johnston, and Snaith 2013; N. G. Park 2020; Saliba, Matsui, Domanski et al. 2016; Saliba, Matsui, Seo, et al. 2016), and are considered to be the fastest advancing solar technology to date. The low-cost and abundance of the materials used in PSCs make them an attractive alternative to traditional solar cell technologies in both residential and industrial settings.

To fit in the perovskite crystalline structure, the organic cations must be small. The most common are methylammonium, ethylammonium and formamidinium, with cesium also being used. The metal cations are divalent metal ions, such as Pb_2^+, Ge_2^+ and Sn_2^+, while the most common halide anions are Cl^- and Br^- and I^- (Saliba, Matsui, Domanski et al. 2016; Saliba, Matsui, Seo et al. 2016). The optical absorption can be tuned with the halide employed, with the iodides resulting in smaller bandgaps and thus, light absorption at larger wavelengths, while the bromides display higher bandgaps and thus, light absorption at shorter wavelengths (Boix et al. 2014; Mosconi et al. 2013). Other advantageous properties include a large absorption coefficient, high charge carrier diffusion length, ambipolar charge mobility and low exciton binding energy (Chen et al. 2019; Ghosh & Singh 2019; Singh and Miyasaka 2016).

A schematic representation of the methylammonium lead iodide ($MAPbI_3$) perovskite ABX_3 structure is presented in Figure 1, which illustrates the organization of the different components in the material.

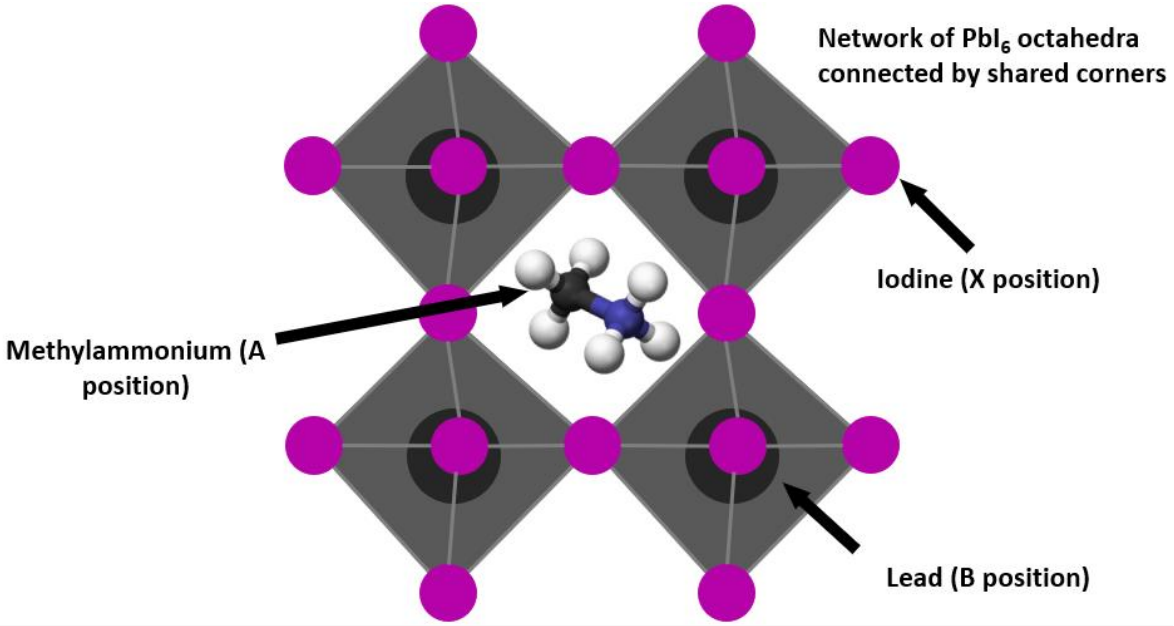

Figure 1: Schematic representation of the perovskite crystal structure of MAPbI3 with respect to the A, B, and X perovskite lattice sites.

Despite significant research efforts, the commercialization of perovskite films has been hindered by some challenges, like the toxicity of the materials and their lack of long-term stability without compromising the power conversion efficiency (Mesquita, Andrade, and Mendes 2018, 2020; Rong, Hu et al. 2018).

In order for PSCs to be commercially viable, they must be able to maintain their stability in a range of environmental conditions, including high temperatures and humidity levels (Burschka et al., 2013). However, the surface of perovskite films is highly sensitive to heat, light, and humidity Mesquita, Andrade, and Mendes, 2018, 2020), which can lead to degradation and reduced performance over time. As a result, researchers have focused on improving the long-term stability of PSCs by controlling grain growth, achieving greater homogeneity, and improving crystallinity (Khassaf et al., 2019; Lohmann et al. 2020).

Diverse efforts have been made to establish techniques for enhancing PSCs. These efforts can be broadly categorized into two approaches: the search for novel perovskite materials that inherently possess stability to degradation and improved performance, and the exploration of fresh preparation methodologies aimed at achieving consistently high-quality durable films (Guo et al. 2023).

Solution based methodologies, mostly spin-coating, are a common and cost-effective way to produce perovskite thin films, particularly on a laboratory scale. Spin-coating involves depositing a perovskite precursor solution onto a substrate and then spinning that substrate at high speeds. The centrifugal force spreads the solution, creating a thin and uniform perovskite layer. After spin coating, the solvent is allowed to evaporate, and a thermal annealing step is often applied to enhance the crystallinity of the perovskite. Other layers on the PSC, like the electron transport layer and hole transport layer, may also be deposited through the same methodology. (Burschka et al. 2013; Im et al. 2014; Saliba, Matsui, Domanski et al. 2016; Saliba, Matsui, Seo et al. 2016).

Other solution-based methodology, like screen printing, slot-die coating, soft-cover coating and spraying coating, are also being applied (Rong, Ming et al. 2018).

However, these methods present challenges, such as the handling of toxic solvents and the difficulty in achieving consistent film formation ,(Liu, Johnston, and Snaith 2013). On the other hand, vacuum processing is a well-established technique in the electronics industry, offering high throughput and reliability, and holds significant potential for commercial scaling-up of perovskite solar cells (Liu, Johnston, and Snaith 2013; Momblona et al. 2016). Furthermore, vacuum deposition eliminates the need for toxic solvents, and enables precise control over film thickness and morphology, leading to improved reproducibility and device performance. Vapor deposition has thus emerged as a promising method for perovskite film fabrication, offering several advantages over traditional solution-based techniques. With the ability to control stoichiometry and crystal size in situ, as well as enabling multilayer deposition and in situ purification, vapor deposition exhibits high reproducibility and reduced risk of contamination (Costa, Azevedo, et al. 2018; Gil-Escrig et al. 2021; Igual-Muñoz, Castillo et al. 2020; Igual-Muñoz, Navarro-Alapont et al. 2020; Momblona et al. 2016; Ono et al. 2016; Zanoni et al. 2023).

Snaith is credited with introducing vapor deposition as a technique to prepare perovskite thin films with improved uniformity compared to spin coating (Liu, Johnston, and Snaith 2013). Among the various device architectures for vapor-deposited perovskite solar cells, p-i-n and n-i-p configurations have emerged as the most promising (Gil-Escrig et al. 2021; Igual-Muñoz, Castillo et al. 2020; Igual-Muñoz, Navarro-Alapont et al. 2020; Momblona et al. 2016; Ono et al. 2016). In these designs, an organic-inorganic perovskite film (i) is sandwiched between two organic semiconductors (OSCs), which consist of a hole (p) and an electron (n) transport layer. OSCs offer several advantages as charge transport layers in multilayer thin-film devices, including structural flexibility, low-temperature processing, and relatively high thermal stability in the glassy state (Momblona et al. 2016).

Bolink's research has achieved the highest power conversion efficiency (PCE) reported to date for fully vacuum-processed PSCs, reaching 20.3% (Momblona et al. 2016). Specifically, a $MAPbI_3$ film sandwiched between two OSCs (C60 and an arylamine derivative) was used.

Several other approaches have been taken to develop methods aimed at enhancing the performance of PSCs. Alternatives to the widely studied $MAPbI_3$ have been considered. For example, $FAPbI_3$ was found to have a narrower and more appropriate bandgap and a superior atmospheric stability. However, it has been reported that it shows less stability in humid environments, when compared to $MAPbI_3$ (Ming Koh et al. 2013). Recently, the effective synthesis of a dual-cation MA/FA mixture has demonstrated that a minor presence of MA prompts the preferential crystallization of FA perovskite into a light-sensitive black phase. This phenomenon results in the creation of compounds and complexes with improved thermal stability and structural integrity, surpassing those achieved with single MA or FA compositions (T.-Y. Yang et al. 2015).

Another key strategy is encapsulation. It involves the implementation of protective layers around the PSC films to shield them from external environmental factors. They act as barriers against moisture and oxygen. They also provide mechanical support to protect the delicate cells from damage. Furthermore, thermal management is a vital aspect of encapsulation. PSCs can experience localized heating, especially during operation under intense sunlight, which may lead

to degradation and performance decline. Encapsulation strategies integrate thermally conductive layers or materials that efficiently dissipate heat, preventing hotspots and ensuring more uniform temperature distribution throughout the device (Dong et al. 2016; Matteocci et al. 2016; Uddin et al. 2019; Weerasinghe et al. 2015). Many materials have been studied as encapsulator layers. These include glass, polymers like PET, PEN, and PDMS, inorganic metal oxides such as Al_2O_3 and SiO_2, epoxy resins for adhesion and protection, and multilayer structures combining organic and inorganic layers. Glass provides a rigid and transparent barrier, while polymers offer flexibility. Epoxy resins provide mechanical strength and adhesion (J. Li et al. 2021; Matteocci et al. 2016; Raman et al. 2021; Uddin et al. 2019).

It has also been reported that the pre-heating of the substrate in solution-based methodology removes residual moisture on the surface and improves the crystallinity of the perovskite (G. Wang et al. 2019; G. Xu et al. 2021).

Anti-solvent engineering is another strategy commonly applied during solution-based methodology. It involves the addition of a non-solvent or a solvent with lower solubility for the perovskite precursor solution. When the anti-solvent is introduced, it rapidly diffuses into the precursor solution, triggering a precipitation process. This results in the formation of perovskite crystals with improved morphology and crystallinity (Xiao et al. 2014).

Furthermore, another strategy applied is the use of additives to stabilize the perovskite, improving its crystallinity and durability. Perovskite thin films can generally incorporate additives through several methods. These include:

1. Addition to the precursor solution during the one-step process.
2. Incorporation into the solution of organic sources during the two-step deposition.
3. Introduction into the antisolvent.
4. Introduction through post-treatment.

The first three methods not only serve to passivate defects but also have an impact on the crystallization process. On the other hand, the final method primarily aims to passivate defects on the interfaces between the perovskite and the hole transport layer/electron transport layer.

Some additives generally applied are Lewis acids, Lewis bases, ammonium salts, lowdimensional perovskites, fullerene-based compounds, quantum dots and, more recently, ionic liquids (S. Liu et al. 2020; Mahapatra et al. 2019; Salim et al. 2021; Xiao et al. 2014; Zhang and Zhu 2020).

Ionic Liquids in Perovskite Solar Cell Engineering

Ionic liquids (ILs) have emerged as promising materials for improving the stability and performance of PSCs. These ionic fluids show promising potential as an additive or as an interfacial layer in PSC due to their advantageous physicochemical properties:

1. **Low volatility:** ILs have low vapor pressure, meaning they do not readily evaporate at normal operating temperatures. This property ensures that the ionic liquid remains stable and does not easily escape from the solar cell, leading to better long-term device stability and reduced degradation (Esperança et al. 2010; Luo et al. 2022; J. Yang et al. 2023).
2. **Good ionic conductivity:** ILs exhibit high ionic conductivity, enabling the efficient transport of ions within the material. This property is essential for proper functioning of the electrolyte or charge transport layers in PSCs. High ionic conductivity facilitates fast

ion diffusion, leading to improved charge extraction, reduced charge recombination, and enhanced overall device performance (Luo et al. 2022; Niu et al. 2021).

3. **High thermal and radiation stability:** ILs possess exceptional stability, making them invaluable in perovskite solar cell applications. They exhibit high thermal stability, enduring elevated temperatures without degradation. Furthermore, ILs demonstrate radiation stability, maintaining their properties when exposed to ionizing radiation. These properties are crucial in solar cells, where the device is exposed to high operating temperatures and solar radiation. These properties ensure the longevity, reliability, and functionality of ILs in PSCs (Deng et al. 2020; Xue et al. 2018).
4. **Wide electrochemical window and chemical stability:** ILs typically have a wide electrochemical window, which refers to the range of voltage or potential within which they remain stable without undergoing undesirable reactions. This property is important in PSCs, ensuring their chemical stability and enabling compatibility with different electrode materials, allowing for efficient charge transfer without significant losses or side reactions (Deng et al. 2020; Fan et al. 2022; Xue et al. 2018).
5. **Tailorability:** One of the key advantages of ILs is their tailorability. The combination of various cations and anions allows for the design and customization of ILs with specific properties. Researchers can choose appropriate ionic liquid compositions to optimize parameters such as viscosity, conductivity, stability, and compatibility with other materials within the perovskite solar cell. This flexibility in tailoring ILs enhances their applicability and effectiveness in specific device configurations (Gu et al. 2022; Pei et al. 2022; Philippi and Welton 2021).
6. **Unusual wetting behavior:** the wetting behavior of an IL refers to its ability to spread and adhere to a solid surface. This allows for an efficient coating and interface layering. Furthermore, this also allows for ILs to fill in and passivate defects on the perovskite structure, which are nonradiative recombination centers, and also promote the perovskite degradation (Borghi and Podestà 2020; Campos et al. 2020; Costa, Coelho et al. 2018; Costa et al. 2023).
7. **Solvating ability:** ILs have the ability to solvate various species, including ions and small molecules. This property allows them to effectively interact with and influence the perovskite materials and other layers within the solar cell. ILs can solvate and stabilize the perovskite precursor materials, promote the formation of high-quality perovskite films, and passivate surface defects or trap sites, thereby improving device performance (Deng et al. 2020; Fan et al. 2022).

Recently, ILs have been explored as interfacial materials in PSCs, where they can optimize the interface contact, improve energy level matching, and suppress ion migration (Hu et al. 2022; Luo et al. 2022; J. Yang et al. 2023; W. Zhang et al. 2020; T. Zhu et al. 2020). They may also increase hydrophobicity, thus inhibiting the decomposition of the device in humid environments (Caliò et al. 2018; J. Wang et al. 2019; W. Zhang et al. 2020). Additionally, ILs have been used as precursors in the solution-based fabrication of perovskite thin films for PSCs applications, assisting in the perovskite crystallization.

Notably, ILs introduced in the interface between the perovskite and the charge transport layers have been shown to decrease the mismatch on the energy levels, improving charge mobility

(Zhou et al. 2023; X. Zhu et al. 2021). A lower charge mobility increases the charge accumulation on the interfaces, and, therefore, the non-radiative recombination rate.

A key challenge for the long-term efficiency and stability of PSCs is the formation of point defects in the perovskite crystal structure, which can further act as centers for charge accumulation and non-radiative recombination. Moreover, they also promote charge scattering, hysteresis effects, and ion migration. These defects inevitably form during the crystal growth of the perovskite films and propagate, promoting the decomposition of the perovskite structure, especially at the grain boundaries. To address this issue, research efforts are focused on developing strategies to passivate the point defects by occupying the defect states with ILs, thereby reducing the non-radiative recombination rate and improving the overall efficiency of the PSCs.

Thus, passivation techniques have become a crucial area of research. Some ILs possess functional groups that can interact with the uncoordinated Pb_2^+ ions, leading to effective passivation of the defects and reducing non-radiative recombination. In addition to their passivating properties, ILs have been shown to enhance the stability of perovskite films by reducing their susceptibility to degradation via oxygen and subsequent reactions under heat and light (Chao et al. 2020; Ghosh and Singh 2019; Hu et al. 2022; Noel et al. 2020; W. Zhang et al. 2020).

Moreover, several other research groups have focused on manipulating the interface and passivating the surface of perovskite solar cells to inhibit ionic migration and charge recombination, ultimately improving the performance and overall stability of the devices (Du et al. 2020; Jiang et al. 2019; Lee et al. 2022; Z. Li et al. 2019; Xiong et al. 2021; Zhao et al. 2018). The incorporation of ILs in these defects has been shown to play a significant role in decreasing the degradation of the perovskite film surface, resulting in enhanced stability and increased efficiency. This highlights the importance of ILs as a powerful tool for passivating defects and improving the quality of perovskite films in the field of photovoltaics.

In recent studies, 1-hexyl-3-methylimidazolium iodide has emerged as a promising IL for optimizing the grain structure of formamidinium lead iodide ($FAPbI_3$) films (Akin et al. 2020). The interaction between the nitrogen in the IL's imidazolium group and the uncoordinated Pb_2^+ has been found to be highly effective at passivating defects and improving the quality of the film. This interaction between the IL and the perovskite film has also been shown to enhance the PSC performance, with a resulting efficiency of 20.6% achieved for the IL-doped PSC compared to 17.1% for the IL-free PSC. In addition, the IL-doped PSC has also shown remarkable stability under challenging conditions. Specifically, the device has maintained >80% efficiency at 60 ± 10% relative humidity and 95% efficiency at 65°C, indicating the effectiveness of the IL in inhibiting degradation and improving the overall durability of the PSC.

Moreover, a slow degradation in the performance of PSCs under continuous radiation for more than 1800 hours at a temperature between 70 and 75 °C has been reported for perovskite - $(FA_{0.83}MA_{0.17})_{0.95}Cs_{0.05}Pb(I_{0.9}Br_{0.1})_3$ - devices incorporating $[C_4C_1im][BF_4]$ applied by solution methodologies (Bai et al. 2019).

Another example of the effectiveness of ILs in passivating perovskite defects is the multifunctional $[PATM][BF_4]$ IL, which has been successfully used to improve the quality and stability of a triple-cation perovskite, $(FA_{0.85}MA_{0.15})_{0.95}Cs_{0.05}Pb(I_{0.85}Br_{0.15})_3$ (T. Li et al. 2021). The IL's amino group coordinated with the Pb^{2+} in the perovskite, reducing the number of charge recombination centers and promoting a more efficient charge transfer. Moreover, a hydrogen bond formed

between the carbonyl group in the IL and the formamidinium in the perovskite, leading to a retardation of film decomposition. These combined effects resulted in a photoelectric conversion efficiency of 21.35% for the IL-doped PSC. The [PATM][BF_4]-doped PSC also demonstrated outstanding stability, with only a 15% decrease in efficiency after exposure to air for 4000 h, under a relative humidity of 40 ± 5%. Furthermore, even under harsher conditions of 85°C for over 1000 h, the efficiency only decreased by 10%. The same decrease was achieved for 800h of full-spectrum sunlight.

Other classes of ILs were employed in the treatment of perovskite solar cell films. A series of ammonium halide salts, based on phenethylammonium iodide with different functional groups attached (−NO_2, −CH_3, and −OCH_3) were employed for post-treatment of perovskite solar cell films (Zhuang et al. 2019). Here both the benzene rings and the ammonium groups were found to coordinate well with the Pb^{2+} ions. The benzene rings in particular exhibited a great passivator behavior due to their electron density. By using the best passivator (containing the −CH_3O group) there were noticeable improvements in various performance metrics of the device. Specifically, the photoluminescence lifetime increased from 0.95 to 2.93 µs, the open-circuit voltage (V_{oc}) increased from 1.11 to 1.18 V, and the power conversion efficiency increased from 19.98% to 22.98%. Additionally, the passivation process helped to reduce the device hysteresis and improve its long-term stability and photostability.

Hydrophobic ILs, like [APMim][PF_6] (J. Wang et al. 2019), [BMPy][TFSI] (Caliò et al. 2018) and [BMMIm][Cl] (W. Zhang et al. 2020) were shown not only to assist on the perovskite crystallization process, but also to provide long-term stability, through the protecting effect of the hydrophobicity of the ILs against atmospheric humidity.

Polymeric ionic liquids have also been applied as passivating molecules for PSCs. PILs have been shown to effectively reduce hysteresis and improve the stability of PSCs against various external degradation factors (Mariotti et al. 2022; S. Wang et al. 2020).

“Solvent-free” vapor deposition methods have been proposed to incorporate stable, high-purity ILs into perovskite thin films (Bai et al. 2019).

However, there are still some issues with the implementation of ILs on PSCs, which hinder its widespread application. There’s a general lack of understanding of the interactions between the ILs and the perovskite and the charge transport layers (Luo et al. 2022; F. Wang et al. 2022). Most studies focus on the overall performance of the solar cells. Further understanding of the interactions at play could be crucial to, taking advantage of the ILs tailorability, design new ILs for these applications. This lack of understanding also implies that leftover residual quantities of ionic liquid on the surface may have unknown effects on the device. There are also difficulties fabricating large areas, hindering the upscaling of the production (Ghosh and Singh 2019; F. Wang et al. 2022).

Furthermore, there are still some concerns regarding the ILs’ properties. Some ILs may still decompose on challenging temperature conditions (Xue et al., 2018), and the hygroscopic nature of many ILs may also be an issue, increasing the decomposition of the water-sensitive perovskites (Fajardo et al. 2017). Moreover, the process of designing new ILs still encounters some challenges, and low cost and easily repeatable synthesis of ILs remains a work in progress (Philippi and Welton 2021).

Conclusion

Perovskite solar cells have emerged as a promising technology for renewable energy generation. However, their lack of stability remains a major challenge that must be addressed to ensure commercial viability. Several studies showed that ILs can be used as additives or interfacial layers in PSCs to optimize interface contact, improve energy level matching, suppress ion migration, and increase hydrophobicity. These properties inhibit the decomposition of the device in challenging conditions and improve the stability and lifetime of PSCs. The potential of ILs as materials for improving the performance and stability of PSCs offers significant implications for the development of low-cost, renewable energy technologies. Further research is needed to fully understand the potential of ILs in PSCs, but the research presented here provides a strong foundation for future investigations. This highlights the importance of exploring new materials and techniques for improving the stability and efficiency of PSCs to accelerate the transition towards a sustainable and cleaner energy future.

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