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Advances and Prospective in Metal Halide Ruddlesen–Popper Perovskite Solar Cells

Shuyan Shao and Maria Antonietta Loi*

Metal halide Ruddlesen–Popper perovskite solar cells (RPPSCs) have attracted a great deal of attention in the research community due to their excellent stability over the 3D counterparts. In 2014, the first RPPSC was reported achieving a power conversion efficiency (PCE) of about 4.7%. To date, this type of solar cells reach a PCE exceeding 18% on lab scale. In this essay, distil strategies to further improve the PCE of RPPSCs are discussed. First, the unique physical properties of RPP are discussed to highlight the importance of film processing and material design, and then the factors that are limiting RPPSCs with special focus on the crystallographic and charge transport properties are addressed. Finally, the opportunities for RPPSCs are discussed, and opinions are provided regarding how to further improve the performance of these devices and on strategies which may advance the technology toward its industrial exploitation.

1. Introduction

Metal halide perovskite solar cells (PSCs) are very promising candidates for commercialization owing to the advantages of easy and low-cost fabrication. 3D metal halide perovskite materials have a ABX_3 crystal structure, in which A is small monovalent ammonium cation such as methylammonium (MA), formamidinium (FA), or cesium occupying the cavity of the inorganic octahedra, B is a divalent metal cation such as lead (Pb) or tin (Sn) sitting in the body centre of the inorganic octahedra, X is a halide anion (Cl^- , Br^- , or I^-) sitting at the corner of the octahedra.^[1–8] These materials have small exciton binding energy (16 meV),^[1] relatively high charge carrier mobility (several tens to hundreds of $cm^2 V^{-1} s^{-1}$),^[2] and long diffusion length ($> 1 \mu m$),^[3] high absorption coefficient and broad absorbing wavelength which ranges in some case into the near infrared region.^[2] These outstanding semiconducting properties has enabled in less than 10 years of work the large international community of researchers working on this class of materials to obtain a power conversion efficiency (PCE) in lab-scale devices of 25.5%, showing an unprecedented fastest progress among all the photovoltaic technologies.^[4,5] However, 3D PSCs often

suffer from performance degradation due to the chemical decomposition of the perovskite crystal structure under environmental stress such as water and temperature variations, which represent a big obstacle toward commercialization.^[6–9]

One of the effective strategies to mitigate this issue is to replace in the perovskite structure, the small hydrophilic organic ammonium cation (MA or FA) by bulky hydrophobic ones such as aliphatic or aromatic mono-, diammonium or triammonium cations (A' , $R-NH_2^+$, $R-(NH_2^+)_2$, or $R-(NH_2^+)_3$). The bulky monoammonium A' spacer cations form interdigitating bilayers via van der Waals forces and turn the 3D into layered Ruddlesden–Popper perovskite (RPP) structure

($A'_2A_{n-1}B_nX_{3n+1}$). While the diammonium spacer cations give rise to structures without van der Waals gap, the cations form hydrogen bonds on both ends with iodine atoms on the adjacent inorganic sheets, yielding to Dion–Jacobson perovskite (DJP) structure ($AA_{n-1}B_nX_{3n+1}$).^[8,10–15] Recently, Kanatzidis et al. reported a new type of layered perovskite ($A'_nB_nX_{3n+1}$), which has a mixed structural feature of the RP and DJP, adopting alternating spacer cations of guanidinium ($C(NH_2)_3$, GA) and MA in the interlayer space (ACI).^[16] In these structures, n represents the thickness of inorganic octahedra sheets sandwiched between the spacer layers in the unit cell.^[2,17–19] It is worth mentioning that a given n represents a specific RP, DJ, or ACI perovskite phase. By simply manipulating the molar ratio of the bulky A' cation to small A cation, n varies from 1 (pure 2D) to ∞ (pure 3D). Thus, the possibility to vary n yields to different perovskite phases with tuneable optoelectronic properties.

The interdigitating monoammonium spacer bilayer results in good self-assemble capability of the RPP layers, which is the driving force for forming highly oriented and crystalline films. It is important to underline that this RP systems are much more thermodynamically stable than the 3D perovskites, which have formation enthalpy (ΔH) strongly positive ($34.50 \pm 1.01 \text{ kJ mol}^{-1}$) for $MAPbI_3$, and weakly positive ($6.69 \pm 1.41 \text{ kJ mol}^{-1}$) for $MAPbBr_3$.^[20] While RP systems have ΔH of formation largely negative ($-49.69 \pm 1.18 \text{ kJ mol}^{-1}$) for low- n compounds ($n = 1–5$) and become positives for high $n > 5$.^[21]

The seminal works, which initiated this research field were published in 2014, when solar cells using quasi-2D RPP phases (low n members, such as 3, 4, or 5), as light absorbing materials with PCEs of about 4.7% were reported.^[12,13] In less than six years, the lab-scale PCE of this type of solar cells has been boosted up to above 18% by manipulating the orientation,

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composition and distribution of the RPP phase as well as the film morphology.^[22] The nominal high n members ($n = 40$ and 60), which actually often form a mixture dominated by a 3D phase and a very small fraction of RPP phase (2D/3D), were also used as light absorbing materials in solar cells, leading to higher efficiency and stability compared to the 3D based counterparts.^[23] The 2D/3D heterostructure perovskites prepared with the deposition at the interface with one of transport layers of the low dimensional system and subsequent deposition of a 3D perovskite or of a RP phases, were also demonstrated to efficient and stable solar cells as compared to the pure 3D phase. These characteristics are mostly due to the effectiveness of the 2D material in passivating the grain boundaries and in encapsulating the perovskite films.^[24–29]

The diammonium cations in the DJP structure not only remove the van der Waals gap, but also reduce the distance between the inorganic layers, which leads to broader light absorption, due to the reduced quantum confinement.^[30] The diammonium spacer interlayer has a drawback the lower degree of conformation freedom, which harms the self-assemble capability of the DJP. Recently, increasing attention has also been drawn to explore the quasi-2D DJP as light harvesting material in PSCs. In 2018, Kanatzidis and co-workers reported the first DJPSC with a PCE of 7.32%.^[30] Recently, they obtained a PCE of 20.6% in DJPSCs using narrow bandgap mixed lead/tin quasi-2D DJP as active layer.^[31]

The large play field represented by the long organic cation offers opportunities to increase further stability and efficiency of PSCs but also to introduce new functionalities in the active layers. In this essay, we will focus the discussions on how to further improve the PCE of the lead iodide based Ruddlesden–Popper perovskite solar cells (RPPSCs). The aim is not much to give an extensive review of the past studies but to provide a critical view of what has been done and propose future research directions. We first discuss the unique properties of the RPP, with emphasis for the structural and charge transport properties, which we believe are central in determining the performance of RPPSCs. We address the challenges faced by the community to control crystallographic and charge transport properties and give our suggestions for new promising research directions which may result in a further improvement of the RPPSCs performance.

2. Unique Properties of RPP and Limiting Factors for Device Performance

2.1. Quantum and Dielectric Confinement in RPP

Due to the quantum confinement (in the inorganic octahedra sheets) and the dielectric mismatch between the inorganic and organic layers, RPP forms natural multiple quantum wells, where the inorganic octahedral layers act as the potential “wells” and the organic insulating A' cations as the potential “barrier” (Figure 1).^[2,32] Density functional theory (DFT) calculations demonstrated that the valence band maximum predominantly consists of hybridized filled metal s-orbital and halide p-orbital, for example: Pb-6s and I-5p orbitals, while the conduction band minimum mainly consists of empty metal p

Figure 1. The energy alignment of the quantum wells with different n value as well as the energy funnelling process.

orbital, that is, Pb-6p orbitals.^[33] Upon photo-excitation, excitons form in the inorganic layers. The bandgap and exciton binding energy decrease with increased inorganic octahedra layer thickness n due to weakened quantum and dielectric confinement effects.^[32] Unlike 3D perovskites, the 2D RPP ($n = 1$) normally has a large exciton binding energy (>170 meV) at room temperature, which cannot be dissociated into free charge carriers by the thermal energy (around 26 meV), shorter carrier diffusion length (≈ 60 nm), and narrower absorption into the visible region ($E_g > 2$ eV). In fact, $n = 2$ phase also possesses similar properties as the $n = 1$ phase.^[12,32] These phases therefore are not ideal as solar cells active layers. In contrast, quasi-2D RPP members with nominal $n = 3, 4$, or 5 , which in reality are often not pure phases but mixed RPP phases with different n members as will be discussed later, have the combined advantages of 2D and 3D perovskites, and have recently become very popular light absorbers in RPPSCs. When n is larger than 7, the RPP phases, also referred to as 3D-like phase, have narrower bandgap, smaller exciton binding energy and high carrier mobility in every direction.^[34] Nevertheless, their structural and thermodynamic stability is inferior to the lower n members (see above). In fact, it becomes more challenging to grow phase-pure with high n numbers due to very similar formation energy. Particularly, RPP with a nominal high n ($\infty > n > 10$) always forms 2D/3D mixtures, where the 3D phase is the dominant component next to a very small fraction of low dimensional phases with different n numbers often located at the interface with the substrate.^[25]

Beside the quantum well thickness n , also the organic spacer cation, namely, its chemical structure, conformation, and dielectric constant (or dipole for a single molecule), plays a crucial role in the structural and optoelectronic properties (bandgap, exciton binding energy, and diffusion length) of the RPP.^[33] Organic spacer cations with higher dielectric constant, for example obtained replacing the aliphatic ammonium by aromatic ammonium, reduce the exciton binding energy and bandgap. Because of their structure these quantum wells have large anisotropic charge transport properties, namely, the

charge transport occurs mostly in the plane where the inorganic octahedra are, while the charge transport from one inorganic layer to another across the insulating A' cations is inhibited or not possible depending to the distance between the inorganics planes.^[33]

Ironically, the unique excitonic and anisotropic charge transport properties of the RPP are among the key limiting factors for the performance of the RPPSCs. However, it is important to underline that quasi-2D RPP members offer important opportunities also in the fabrication of efficient and color pure light emitting diodes.^[35,36] In the following sections we shall focus the discussions on how to use the opportunity offered by this class of materials and how advanced processing technique and rational material design can be used to obtain efficient and stable RPPSCs.

2.2. The Problem of the Orientation

Considering the vertical sandwich-like structure of the solar cells (Figure 2a), RPP films with inorganic layers perpendicular to the substrate are desired to facilitate the charge transport to the electrodes (Figure 2b). RPP films with randomly and parallel oriented inorganic layers, will hinder charge transport in the direction perpendicular to the substrate, causing severe recombination from free electron and holes, which results in solar cells with limited current density and fill factor.

The orientation of the RPP layers depends on the processing conditions such as deposition methods, annealing timing, use of additives and solvents, interaction with the layer where they are deposited.^[37–39] In the method that has become conventional in this field, the crystallization occurs when the film is spin dry at room temperature from a precursor solution using DMF as solvent, yielding randomly oriented RPP layers. So far, 2-phenylethylammonium ($\text{C}_6\text{H}_5\text{C}_2\text{H}_4\text{NH}_3^+$, PEA^+) and *n*-butylammonium ($\text{n-C}_4\text{H}_9\text{NH}_3^+$, BA^+) have been the most extensively used organic spacer A' cations.^[12–14] The pioneering works of 2014 on RPPSCs employing these two spacer cations yielded a PCE of 4.73% and 4%, respectively.^[12,13] These early devices suffered from low current density and FF, most probably caused by the poor charge transport due to the imperfect alignment of the inorganic layers respect to the electrodes.

In 2016, Tsai et al., successfully realized vertically oriented inorganic layers in $\text{BA}_2\text{MA}_3\text{Pb}_4\text{I}_{13}$ RPP film by employing a so-called hot-casting method. The improved charge transport in

between the electrodes of the solar cells gave rise to a remarkable improvement in the current density and FF, leading to a PCE of 12%.^[37] These results demonstrated to the scientific community the importance of the alignment of the inorganic planes respect to the substrate in this highly anisotropic material. Interestingly, the addition of Cesium cations, chlorine based additives and the use of DMSO as solvent were also demonstrated to induce the grow of such vertically oriented RPP layers.^[40]

Tuning the chemical structure of the A' organic spacer cation and manipulating surface chemistry of the bottom layers are potential alternative strategies to manipulating the orientation of RPP layers. However, compared to the bulk of works devoted on developing new processing strategies, studies investigating the effect of the chemical structure of the organic spacer cations on the orientation of RPP and the interaction with the material of the bottom layer, have been very limited. DFT calculations demonstrated that the packing geometry and orientational disorder of the bulky organic spacer cations determine the formation energy and the orientation of the inorganic layers.^[41] For example, the organic cation such as 2-fluorophenethylammonium (oF1PEA) with high orientational disorder and edge to edge packing geometry leads to randomly orientated inorganic layers and poor charge transport.^[41] Instead, inorganic layers become perpendicular to the substrate in the RPP films using organic cations such as PEA, 3-fluorophenethylammonium (mF1PEA), or 4-fluorophenethylammonium (pF1PEA) with edge to face packing geometry and low orientational disorder. Interestingly, butylammonium ions with a shorter branched-chain (iso- BA^+) have been found to yield more crystalline and vertically oriented inorganic layers than the linear analogues, with significant consequences for the device functioning.^[42]

Despite the progress discussed above, the link between the processing methods, additives, chemical structure of the organic spacer cation, the interaction with the bottom layer and the orientation of the RPP layers is still unclear. Therefore, gaining insight into the mechanism of the orientation of RPP and the parameters which govern the crystallization dynamics has great implications for further improving the performance of the solar cells. However, these subjects have seldomly been investigated until very recently. In situ grazing-incidence wide angle X-ray scattering experiments revealed that the heterogeneous nucleation and growth of the RPP occurring at the liquid–air interface is responsible for the oriented layers and the high crystallinity of the films, while the nucleation and crystallization taking place in the bulk of the solution is responsible for the random orientation of the RPP.^[43,39] In order to perfect the orientation of the RPP, strategies that can effectively suppress the bulk crystallization process shall be developed.

2.3. The Conductivity Perpendicular to the Inorganic Sheets

Since it is very challenging to grow perfectly vertically oriented inorganic layers, improving the intrinsic conductivity perpendicular to the inorganic layers across the organic spacer cations (out-of-plane) could be an alternative strategy to facilitate

Figure 2. a) Typical solar cell structure consisting of anode, hole transport layer, RPP, ETL, and cathode. b) the parallel and vertical orientation of the RPP.

the charge transport in RP solar cells. So far, very limited research activities have been carried out to enhance the intrinsic out-of-plane conductivity of RPP materials. Charge carrier tunnelling through the organic spacer layers, which is determined by the inorganic-to-inorganic interlayer spacing, intra and intermolecular electronic coupling of the spacer cations as well as tuning of the barrier height of the quantum well, limits the out-of-plane conductivity in these systems. Reducing the inter-layer spacing by using an A' cation, which has shorter chain length or a less extended structural conformation enabling longer distance between ammonium cation and the iodine of the inorganic layer, reinforcing intramolecular or intermolecular electronic coupling such as introducing hydrogen bond and π - π stacking interactions of the organic spacer cations as well as improving energy alignment between the organic spacer cation and inorganic layers are among strategies that may result effectively in improving the out-of-plane conductivity. Therefore, tailoring the chemical structure of the A' spacer cation is an important strategy to pursue in the future.

For example, Liu et al., used n-propylammonium cation (PA⁺) with a shorter alkyl chain length to prepare PA₂MA₄Pb₅I₁₆ films for better charge transport and extraction properties.^[44] Stupp et al., reported a series of $n = 1$ layered perovskites with the form (aromatic-O-linker-NH₃)₂PbI₄, where the naphthalene, pyrene, or perylene is used as the aromatic moiety and ethyl, propyl, or butyl is used as linker.^[45] Generally, these materials have superior out-of-plane conductivity compared to the control RPP materials using aliphatic spacer cations due to stronger electronic coupling between the aromatic core of the organic spacer cations. Their out-of-plane conductivity decrease with the linker length, whereas the optical bandgap and barrier height of the quantum well show an opposite trend. It is worth mentioning that the structural conformation also plays an important role in the out-of-plane conductivity. Enlarging the distance between the ammonium cation and the negatively charge iodine (d_{N-I}) of the inorganic layer by forming intramolecular hydrogen bond with the pyrene organic spacer enhances the out-of-plane conductivity compared to the counterpart with smaller d_{N-I} adopting the extended conformation. These seem to be a new strategy to manipulate the optoelectronic properties of the RPP materials by considering both the energy levels of the organic spacer and the distortions induced by the spacer in the inorganic layer.^[45] While the approach appears promising only devices with efficiency of 1.38% were obtained with $n = 1$. Barea et al., introduced conjugated organic spacer cation anilinium iodide (C₆H₅NH₃I) in RPP (C₆H₅NH₃I)₂(CH₃NH₃I) _{$n-1$} (PbI₂) _{n} (n is from 1 to 5).^[46] These RPP have superior charge transport compared to the BA based counterparts due to stronger electronic coupling of the anilinium spacer cations. Interestingly, the excitons in C₆H₅NH₃I based $n \geq 2$ films are readily ionized into free carriers at room temperature. Consequently, the C₆H₅NH₃I based $n = 2$ phase enables similar J_{SC} as the $n = 3$ and 4 phases. These findings open a potential research direction toward more efficient RPPSCs. Finally, it is important to underline that besides the promise of an increased transport it is not given that by changing the nature of the A' cation the thermodynamic stability will be not detrimentally affected, therefore the new system should always be tested for stability.

2.4. Phase Distribution/Segregation of RPP and Energy Loss

At this point it is important to underline that is very challenging to grow pure and single phase $n > 1$ RPP thin films due to the small difference in the formation energy of RPP of neighbouring n values in the conventional fast crystallization process. Furthermore, as mentioned earlier this difficulty grows with the increase of the n number.

Due to negative enthalpy of formation (ΔH), the small RPP n ($n \leq 5$) phases are energetically more favourable to form than $n > 5$ phases.^[34] Generally, this low n phases are oriented with the inorganic layers parallel respect to the substrate hindering the charge transport in RPPSCs.

While energy funnelling may play an important role in converting excitons generated in the low n phases into free charge carriers in high n phases (Figure 1a).^[47] The efficiency of the energy transfer and charge transport highly depend on the distribution, dimension and composition of these RPP phases. A vertical phase distribution with a cascade from well aligned low n , to high n phases should form from top to the bottom of the film to facilitate the energy funnelling process and enhance the resistance to moisture. Moreover, the lateral phase distribution is also critical for the energy transfer and charge transport.^[47] Large quantity and dimension of parallelly oriented $n = 2$ domains were proven, by confocal photoluminescence measurements, to be unfavourable for exciton diffusion and subsequent energy transfer (Figure 3a).^[14] Moreover, these parallel oriented $n = 2$ phase hinders the charge transport and blocks the charge collection at the anode interfaces (Figure 3c).

Figure 3. Schematic illustration of the phase distribution and segregation in an RPP film grown by a) conventional method, b) vacuum method. Reproduced with permission.^[14] Copyright 2019, American Chemical Society.

Consequently, severe second order recombination occurs in solar cells, leading to low current density and FF. The quantity and the size of the segregated domains of $n \leq 2$ phases should be minimized to facilitate energy funnelling and charge transport (Figure 3b,d).^[14] Charge transfer, where electrons transfer from low n to high n phases, and holes transfer from high n to low n phases, was also proposed to account for the mechanism of charge separation in RPPSCs.^[48] We would like to point out that significant energy loss occurs during the energy funnelling/charge transfer from high energy hot or ground state excitons to lower energy states. Extracting these high energy charge carriers to minimize the energy loss is a big challenge to be tackled. Regardless of the debate about the mechanism, opinions on how to obtain the favourable phase distribution and composition for higher PCE of the RPPSCs converge.

Processing methods that slows down the crystallization process, are very effective in eliminating $n \leq 2$ phases. For example, the addition of DMSO to the precursors solvent reduces the quantity of the parallel oriented $n \leq 2$ phase in the RPP film, improving the charge transport in solar cells.^[49] It has also been demonstrated that a post-deposition treatment in vacuum considerably reduces the quantity and dimension of segregated $n \leq 2$ domains, facilitating energy funnelling from low, to high n phase and charge transport in the active layer (Figure 3c).^[14] As a consequence of these improved spatial distribution and smaller dimension of domains, RPPSCs fabricated with this method exhibit a threefold improvement in the PCE.

The chemical structure of the organic ammonium cations may also affect phase distribution in RPP film. The RPP film using oF1PEA as organic spacer cation has shown unfavourable random phase distribution compared to favourable vertical phase distribution in RPP films based on PEA, mF1PEA, and pF1PEA cations, hindering the energy funnelling between the different RPP phases.^[41]

It is worth mentioning that so far, relevant research works provide mostly qualitative analysis of the effect of the phase distribution on the charge transport, energy funnelling/charge transfer and solar cell performance. The effect of the orientation disorder and segregation domain size of the RPP on the exciton diffusion and energy funnelling/charge transfer efficiency have been overlooked. Simulations could help to establish a model to quantify the effect of the phase orientation, distribution, and domain size on relevant physical properties and finally on the performance of the RPPSCs. The energy loss happened due to energy tunneling or charge transfer from the hot/ground state excitons to lower energy states is another open topic to be addressed in the future studies.

So far, only a handful of papers reported phase-pure $n > 1$ RPP thin films by melt-processing, hot casting, or using as molten salt spacer n -butylamine acetate.^[37,50–52] However, only two examples are reported of phase-pure RPP thin films used to fabricate RPPSCs, which exhibit PCEs lower than 17%, therefore slightly inferior to state-of-the-art RPPSCs containing multiple RPP phases (>18%). Since $n = 3, 4$, and 5 phases show exciton binding energy much larger than the thermal energy, one of the puzzles needs to be solved is how excitons will dissociate into free charge carriers in those phase-pure RPP films. The typical solution in case of excitonic materials, is the design of type two heterostructures for exciton splitting, obviously if the exciton

diffusion length is sufficient to guarantee also an active layer thickness that allows sufficient absorption. However, a recent study suggested that the excitons in $n > 2$ phases dissociate into free charge carriers at the low energy edge states,^[53] showing a similar energy loss to the one occurred during the energy funnelling/charge transfer process between different RPP phases.

Another recent study indicated that phase-pure RPP film has superior charge transport properties compared to the multiple-phase RPP film.^[50] It is however difficult to pinpoint the role of the phase purity in the charge transport properties due to the difficulty of controlling the orientation of the inorganic layers and film morphology.

Concluding, phase pure materials would need first to be controlled and to be carefully studied to understand their real potential for optoelectronic devices.

2.5. Manipulating Exciton Binding Energy and Diffusion Length

Since excitons need to dissociate into free electron and holes to contribute to the photocurrent of solar cells, the reduction of their binding energy and the increase of their diffusion length may help to improve the charge separation and collection of the RPP solar cells. Increasing the dielectric constant of the organic spacer is one of the effective strategies to reduce the exciton binding energy of the RPP.^[33] For example, RPPs using aromatic spacer cations have smaller exciton binding energy compared to that using aliphatic spacer cations.^[33] Recent DFT calculation performed by Grozema and coworkers demonstrated that introducing extra functionality into the organic spacer cations such as electron donating or withdrawing groups allows the formation of localized states either in the organic or the inorganic part of the 2D perovskite.^[54] Furthermore, their results indicated the feasibility of tuning independently the energy of the bands located in the organic spacer by manipulating its electron withdrawing/donating nature without affecting the band structure of the inorganic part (of course if the inorganic octahedra are not distorted in their assembly by the organic moiety (vide infra)). Very recently, the authors from the same research group experimentally demonstrated that mobile free charge carriers form with ten times higher yield by incorporating a strongly electron accepting moiety, such as perylene diimide, on the surface of the 2D perovskite nanoplatelets as compared to the one without the electron accepting moiety.^[33]

Alternatively, the diffusion length of the excitons has also an effect in the charge carrier separation and collection of RPPSCs. The organic spacer cation has a significant impact on lattice distortion, which influences the effective mass, the dynamics and diffusion of the excitons in the RPP phase. Increasing the cross-sectional area of the organic spacer causes distortion of the inorganic lattice, and a reduction in the orbital overlap between neighboring octahedra, which increases the effective mass of excitons and slow down their diffusion.^[33] However, the effective mass model does not explain the higher diffusivity of excitons in PEA_2PbI_4 as compared to those in BA_2PbI_4 .^[55] Prins and coworkers attributed the higher diffusivity in PEA_2PbI_4 to its higher lattice stiffness.^[55] Their results indicated the formation of exciton-polarons with larger effective mass and a lower

diffusivity and show a dominant role of exciton–phonon coupling in the spatial dynamics of the excitons in 2D RPP.

Therefore, the structural rigidity could be used as one of the parameters to design and optimize RPP with suitable optoelectronic properties. However, other possible factors that could contribute to the difference in diffusivity of excitons of these two RPP structures such as the dielectric constant of the organic spacer cations were not addressed. Even though it is generally accepted that the aromatic spacer cations have higher dielectric constants than the aliphatic counterparts, accurate determination of the dielectric constant values of the two RPP structures should be determined. Moreover, both theoretical and experimental results from previous works present controversial and even contradictory values and variation trends in the reduced exciton mass of the RPP materials, such as PEA_2PbI_4 and BA_2PbI_4 , which poses more challenges to accurately pinpoint the factors/parameters determining the exciton diffusion behavior.^[44,45] Therefore, future studies need to not only address how to obtain accurate and reliable physical quantities such as dielectric constant, structural distortion, and effective mass of the excitons, but also develop a more quantitative and accurate theoretical model that can predicts and explain the different excitons diffusivity in RPP using various organic spacer cations. Previous research works investigating the temperature dependence of the photo-conductivity of the halide perovskite materials by terahertz spectroscopy highlight that the electron–phonon scattering fundamentally sets the upper limit of the free charge carrier mobility in 3D perovskites.^[56] This means that the impact of the organic cations such as cross section area, intra-molecular binding or interactions and structural rigidity, on the free charge carrier mobility of the low dimensional perovskite phases should also be taken into account.

In short, recent studies offer very promising new approaches for tuning the optoelectronic properties of these materials to develop stable RPP phases with improved charge separation for solar cells applications. As the relevant studies are still at early stage and mostly investigating spectroscopic properties, investigations reporting the relationship between the structure of the organic spacer, exciton/free charge carrier transport properties and the performance of RPPSCs are still missing, opening potential new research directions for the RPPSCs community.

2.6. The RPP Film Morphology

The film morphology, such as grain boundaries and surface coverage, is another important factor determining the performance of any metal halide perovskite-based device.

Grain boundaries, are the location where structural defects form, and are prone to trap free charge carriers and cause non-radiative trap-assisted charge recombination, leading to solar cells of low open circuit voltage.^[57–60] Moreover, these structural defects may also trap excitons and cause their recombination before their separation in electrons and holes pairs, reducing the photocurrent of the solar cells. Furthermore, pin holes may cause high leakage current by forming shunts, which also reduce the V_{OC} of the solar cells. Therefore, RPP film with compact morphology and big grains are desirable to achieve highly efficient solar cells.^[48]

The film morphology is determined by the crystallization dynamics of the RPP film, which is extremely different from the

one of the 3D metal halide perovskites. It has been shown that slowing down the crystallization process by adding, for example, Cs cation, chlorine based additive, DMSO solvent, and using a vacuum method not only improves the crystallinity, but also the morphology of the RPP film, reducing the trap density significantly.^[13,30,33] Furthermore, the manipulation of the chemical structure or composition (using a mixture of different cations such as A' and A) plays an important role in the RPP film morphology. For instance, replacing 20% MA cation by FA cation significantly enlarges the crystal grains from several hundred nanometres to several micrometres, and produces compact film morphology without any cracks,^[49] and consequently a reduced trap density. Replacing 25% PEA by perfluorinated phenethylammonium (5F-PEA) helps to eliminate the pin holes, at the same time keeping the vertical orientation of the RPP film.^[61]

To summarizing the processing method, the chemical structures/compositions of the organic cations affect the crystallographic, morphological, optical, and electronic properties of the RPP, which are key factors determining the efficiency of the RPPSCs.

3. Challenges and Outlook

The PCE and stability of RPPSCs are far from performance of the 3D perovskite and still not sufficient for commercial applications, even if their higher thermodynamic stability is of enormous importance for applications. To further advance this type of solar cells, future research directions need to focus on the control of the different phases of RPP, with special attention in maintaining the thermodynamic stability as focus of the investigation as some composition with enhanced device performances may have lower stability than the 3D perovskite devices. Solution processing poses challenges in control the crystallization dynamics to fully eliminate the crystallographic and phase disorder of the RPP film. This problem will be amplified when scalable techniques will be used instead of spin coating. Here we would like to propose a more careful look at some recent and less recent literature that have been using Langmuir–Blodgett as technique to control RPP phases.^[62,63] The RP perovskite materials are ideal systems for the Langmuir–Blodgett technique, opening the opportunity to obtain pure phases such as $n = 5$ or larger.

The vast structural diversity and tuneability of the organic spacer cations could potentially offer the opportunity for highly efficient and stable RPPSCs. The organic spacer cations, such as its cross section area, conformation, hydrophobicity, and the van der waals force between them, are among the key factors determining the structural stability of the RPP. This indicates the potential research directions to improve the stability of the RPP. However, the understanding of how the chemical structure of the organic spacer cations influences optical, electrical properties, and thermodynamics of the RPP is still very limited, and much more theoretical and experimental studies would be needed to understand this influence and be able to design new materials with the necessary properties for efficient and stable solar cells.

When discussing the phase stability of these system a large role is played by the transport layer in which the material is deposited, as its surface energy influences the nucleation of the RPP. However, the role of the transport layer is fundamental in the device functioning and more attention should be devoted to

high quality hole and electron transport layers (ETL) that can improve the anode/perovskite and cathode/perovskite interfaces, to further improve not only the efficiency but also the stability of the RPPSCs.

Beside the efficiency and stability issues, research activities to explore the viability of the large area RPPSCs using processing techniques compatible with massive production are also imperative to push this technology closer to practical applications. Since the crystallization dynamics is sensitive to the processing conditions, we foresee new challenges in getting RPP film with uniform orientation, phase distribution and composition by large scale processing techniques. The future will tell if roll-to-roll Langmuir–Blodgett will be the answer to make of RPP solar cells the new big thing in photovoltaic.

4. Conclusions

In conclusion, we discussed the unique properties of the RPP, and the factors limiting the performance of solar cells fabricated with these systems. We spot as main issues the difficulty to control the phases of RPPs. We reviewed briefly the past research activities, without intention to be exhaustive, on how to improve the PCE of the RPPSCs by optimizing the film processing and material chemical structure. We believe a comprehensive and collaborative investigation on the fundamental physical and chemical properties of the RPPs involving experimentalist and theorist is imperative to exploit the large potential of this class of materials obtaining not only more efficient and stable solar cells but also new functionalities by exploiting the tunability of organic molecules. The development of a large scale deposition technique that can allow controlling different RP phases is pivotal to advance this research field.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

charge carrier transport, crystallographic properties, phase distribution, Ruddlesden–Popper, perovskite solar cells

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