Drop-Casting Method to Screen Ruddlesden–Popper Perovskite Formulations for Use in Solar Cells

Chuantian Zuo, Andrew D. Scully, and Mei Gao*

ABSTRACT: Small-area metal–halide perovskite solar cells (PSCs) having power-conversion efficiencies (PCEs) of greater than 25% can be prepared by using a spin-coated perovskite layer, but this technique is not readily transferrable to large-scale manufacturing. Drop-casting is a simple alternative method for film formation that is more closely aligned to industry-relevant coating processes. In the present work, drop-casting was used to prepare films for screening two-dimensional Ruddlesden–Popper (2DRP) metal–halide perovskite formulations for potential utility in PSCs, without additional processing steps such as inert-gas blowing or application of antisolvent. The composition of the 2DRP formulation used for drop-casting was found to have a profound effect on optical, spectroscopic, morphological, and phase-distribution properties of the films as well as the photovoltaic performance of related PSC devices. This facile method for screening film quality greatly assists in speeding up the identification of perovskite formulations of interest. The optimal 2DRP perovskite formulation identified from screening was utilized for industry-relevant one-step roll-to-roll slot-die coating on a flexible plastic substrate, producing PSCs having PCEs of up to 8.8%. A mechanism describing film formation and phase distribution in the films is also proposed.

KEYWORDS: drop-casting, self-spreading, slot-die coating, two-dimensional perovskite, printable perovskite solar cells

INTRODUCTION

Inorganic–organic metal–halide perovskite solar cells (PSCs) are promising candidates for future use in photovoltaic devices due to their low cost, low processing temperature, and high power-conversion efficiency (PCE). Various solution-deposition methods are available for fabrication of the perovskite layer, and optimization of these methods is critical for obtaining high-performance PSCs. Examples of methods used for lab-scale solution deposition of perovskite layers for PSCs are illustrated in Figure 1.

Spin-coating is the most commonly used laboratory method for producing the perovskite layer and has resulted in PSCs having PCEs of more than 25%. It is a conveniently simple approach for fabricating lab-scale devices but results in most of the solution being spun off the substrate leading to substantial material waste. It is also impractical for achieving uniform large-area films. Furthermore, the drying kinetics of spin-coated solutions is much faster than other conventional industrial coating methods due to the combination of centrifugal force and gas flow caused by the spinning substrate, making the direct translation of spin-coated perovskite formulations to large-scale coating methods extremely difficult. Consequently, spin-coating is generally considered unsuitable for producing large-area PSC devices at an industrial scale.

A variety of spin-coating-free methods have been developed to prepare PSCs, including vapor deposition, screen printing, blade coating, slot-die coating, and meniscus-assisted solution printing. An advantage of the vapor deposition method is that it avoids the use of solvent, but it...
Perovskite film formation behavior observed in the drop-casting method can provide a good indication of film formation by using the more technically complex slot-die coating method. For example, we previously found very little difference in the average PCE of PSC devices comprising a perovskite layer formed by drop-casting or slot-die coating the two-dimensional Ruddlesden–Popper (2DRP) perovskite formulation BA$_2$MA$_3$Pb$_4$I$_{13}$ (BA = n-butylationmmonium cation, MA = methylammonium cation).

Because the performance of PSCs comprising a slot-die-coated perovskite layer is highly sensitive to a variety of parameters, such as perovskite composition, solvent, additives, and coating conditions, the comparative simplicity of the drop-cast method offers an attractive means of simplifying and accelerating the process of identifying optimal perovskite formulations and coating conditions. Identifying further perovskite formulations suitable for slot-die coating will provide greater choice and better performance for the wide variety of potential end-use applications of PSC technology.

Herein, we report the results of a systematic approach for screening an array of 2D organic–inorganic hybrid lead–halide 2DRP perovskite formulations comprising a set of straight-chain alkylammonium cations, based on analyzing simple drop-cast films by using routine optical, spectroscopic, microscopic, and electrical techniques. This class of perovskites is of interest due to their superior environmental stability and unique optoelectronic properties. This screening process resulted in a substantially smaller subset of formulations of potential interest, with the reliability of the screening verified by measurements of device PCE. A film-formation mechanism is proposed that is consistent with the observed film morphologies and phase distributions. The perovskite formulation having the highest PCE determined from the morphologies and phase distributions. The perovskite composition, solvents, and additives all have a strong influence on the crystallization process and, ultimately, film quality. Acceleration of the upscaling of PSC technology requires the development of a facile method through which suitability of potential materials and preparation conditions for large-scale coating can be assessed quickly.

Identifying suitable perovskite materials that form high-quality films via natural drying requires substantial effort due to the wide variety of available perovskite compositions, including inorganic perovskites,$^{17}$ mixed-cation perovskites,$^{18}$ mixed tin/lead perovskites,$^{19}$ and 2D/3D perovskites.$^{20–23}$ The perovskite composition, solvents, and additives all have a strong influence on the crystallization process and, ultimately, film quality. Acceleration of the upscaling of PSC technology requires the development of a facile method through which suitability of potential materials and preparation conditions for large-scale coating can be assessed quickly.

Recently, we reported a drop-casting method to prepare inorganic–organic hybrid lead–halide perovskite films for PSCs$^{24–29}$ where the perovskite solution spreads spontaneously on a heated substrate to form uniformly smooth, high-quality perovskite films upon drying. The self-spreading in the drop-casting method is caused by the unbalanced surface tension of solution/substrate, solution/air, and air/substrate interfaces, enabling film preparation without assistance of depositing apparatus.$^{29}$ Apart from conserving time and materials, the drop-casting method is, in principle, much more like an industry-relevant coating processes, especially slot-die coating, than spin-coating. The similarity in the fluid dynamics and drying kinetics of films produced by using the drop-cast and slot-die methods from a given precursor formulation$^{24}$ means that the film formation behavior observed in the drop-casting method can provide a good indication of film formation by using the more technically complex slot-die coating method.

**Solution Preparation.** The MAPbI$_3$ precursor solution was prepared by dissolving 0.3 M PbI$_2$ (Alfa Aesar) and 0.3 M MAI (Greatcell Solar) in dimethylformamide (DMF) (Sigma-Aldrich). Precursor solutions for 2D perovskites X$_m$MA$_n$Pb$_{4m}$I$_{13n}$ with concentrations of 0.3 M of Pb$^{2+}$ were prepared in DMF. The molar ratio of the reagents PbI$_2$:MAI = C$_n$A$_m$ was 4:3:2. Precursor solutions for (C$_4$A)$_3$MA$_2$:Pb$_{4}$I$_{13}$ perovskites were prepared by fixing the Pb$^{2+}$ concentration at 0.3 M; the concentrations of MAI and (C$_4$A)$_3$Cl can be calculated by using the n value. The precursor solutions were stirred at 70 °C for 1 h. The PEDOT:PSS solution for the R2R slot-die process was made by mixing 1 mL of PEDOT:PSS (HC Starck, Baytron P Al 4083), 1 mL of deionized water, and 1.6 mL of isopropanol.

**Materials Characterization.** Photographs of the films were taken with an Epson Perfection V700 photoscanner. The SEM images were taken with a Zeiss Merlin field emission SEM (FE-SEM) operated at 5 kV. Optical microscopy images were taken by a LW200-3JT microscope. UV–vis absorption spectra were recorded by a Lambda 35 PerkinElmer absorption spectrometer.

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Steady-state PL emission spectra were recorded by a luminescence spectrometer (Edinburgh Instruments, FLSP920).

**Device Fabrication Using the Drop-Casting Method.** The perovskite and charge transport layers were prepared in air with humidity between 30% and 50%. Patterned ITO glass was cleaned in detergent (Deconex12PA detergent solution), deionized water, acetone, and isopropanol sequentially by ultrasonication and then treated with UV-ozone for 15 min. PEDOT:PSS was spin-coated at 5000 rpm for 30 s. The substrate was then heated on a hot plate at 150 °C for 10 min in air. After cooling to room temperature, the substrate was heated on a 60 °C hot plate for 2 min, and then 6 μL of perovskite solutions was dropped onto the center of the 25 mm × 25 mm substrate. The solution spontaneously spread on the substrate and dried in 1 min, forming a round black film. The substrate was then heated at 100 °C for 2 min. PC61BM in chloroform with a concentration of 10 mg mL\(^{-1}\) was spin-coated onto the perovskite layer at 1000 rpm for 30 s. Then PEIE (0.05% w/w in isopropanol) was spin-coated onto the PC61BM layer at 4000 rpm for 30 s. Finally, 100 nm Ag was evaporated through a shadow mask to give an active area of 0.1 cm\(^2\).

**Device Fabrication Using the Roll-to-Roll Process.** The setup of the slot-die roll-to-roll coating system and the coating of the PEDOT:PSS layer can be found in our previous work. A \((C_2A)_{2}(MA)\), PbI\(_{16}\) precursor solution containing 0.6 M Pb\(^{2+}\) was used. The speed of the substrate movement was 0.5 m min\(^{-1}\), and the solution feed speed was 30 μL min\(^{-1}\). The coated film was passed over the first curved hot plate at 60 °C to dry the film, and then over a second hot plate at 100 °C, before being collected in a roll. After completion of the perovskite layer, the substrate was cut into 25 mm × 25 mm to make solar cells. PC61BM, PEIE, and Ag layers were deposited as described above.

**Device Measurements.** The J–V curves were measured under an inert atmosphere by using a Keithley 2400 source meter under standard solar irradiation (AM 1.5G, 100 mW cm\(^{-2}\)). The light intensity was calibrated by using a reference cell (Hamamatsu S1133 with KG5 filter, 2.8 × 2.4 mm\(^2\) of photosensitive area), which was calibrated by a certified reference cell (PV Measurements, certified by NREL) under 1000 W m\(^{-2}\) AM 1.5G illumination from an Oriel AAA solar simulator fitted with a 1000 W Xe lamp.

## RESULTS AND DISCUSSION

The size of the bulky organic cations (typically alkylammonium cations), R, and/or the value of n, in 2DRP perovskite compositions, R\(_m\)MA\(_{m-1}\)Pb\(_m\)I\(_{3n+1}\), have been shown to have a major influence on the optical and optoelectronic properties of spin-cast 2DRP perovskite films. This is reflected in the crystal growth and orientation, passivation of trap states, and hindering of ion motion. Identifying the combination of size of R and value of n giving optimal perovskite film properties for PSCs requires the screening of the potentially large number of 2DRP perovskite formulations generated by systematically changing the cation size and n value. In this work, the cation size was varied through the use of straight-chain alkylammonium ions having different chain lengths. For this purpose, a set of the 2DRP perovskite formulations \((C_2A)_{2}MA_{m-1}Pb_{m}\) were prepared by using dimethylformamide (DMF) as the solvent, where MA is the methylammonium cation and C\(_2A\) represents a homologous series of linear straight-chain alkylammonium cations with m being the number of carbon atoms in alkyl chain. The alkylammonium salts used in this work, and their abbreviations, are shown in Figure 2. When the carbon chains in C\(_2A\) are longer than m = 6, charge transport in the 2DRP perovskite is hindered due to the insulating property of the carbon chains, which may lead to inferior photovoltaic performance. If n is too large, the concentration of 2DRP phases in the film becomes much lower than the 3D perovskite phase, reducing the potential beneficial effects of the 2D phases. Consequently, the array of 2DRP formulations studied in this work was restricted to those having m ≤ 6 and n ≤ 8.

Drop-casting provides a straightforward means for rapidly preparing films from 2DRP perovskite formulations in the required parameter space. The images of the drop-cast perovskite films prepared from the set of precursor solution compositions shown in Figure 3a illustrate the significant influence of the solution formulation on the visual appearance of the dry films. Considering initially the top row of images in Figure 3a, the film formed by using the \(\{m = 2, n = 4\}\) formulation is a hazy, gray color with a very rough and nonuniform surface, similar to the 3D MAPbI\(_3\) film. The significant light scattering by this film is the origin of the elevated baseline in its UV–vis absorption spectrum, where the “baseline” is taken here to be the absorbance at 800 nm which is beyond the bandgap of 3D perovskite (Figure 3b). In contrast, the brown films prepared from the \(\{m = 3\text{--}5, n = 4\}\) formulations are relatively glossy with minimal haze as seen in the lower baselines in their UV–vis absorption spectra. Uniform, compact crystalline grains in the perovskite layer are a prerequisite for a PSC device to display good photovoltaic performance, and these physical properties typically yield this type of glossy film showing a high degree of specular reflectance. The brown film obtained by using the \(\{m = 6, n = 4\}\) formulation has a high degree of haze, as seen in its elevated UV–vis absorption baseline.

The strong dependence of perovskite film color on n seen in the second row of images in Figure 3a arises from contributions to optical absorbance by constituent perovskite phases having different dimensionalities as defined by the number of Pb–I octahedron layers in their crystal structure. The bandgap of the perovskite phases decreases (red-shifts) with increasing dimensionality. The multitude of bands in the UV–vis absorption spectra of the drop-cast films (Figures 3b,c) show that perovskite phases having a variety of dimensionality exist in the films. The glossy film obtained by using the \(\{m = 4, n = 1\}\) formulation shows high specular reflectance implying uniform, compact grains and displays one sharp peak associated with a phase consisting of a single layer of Pb–I octahedrons; the phase purity of this film is expected because no other phases are stoichiometrically possible in a solution of this formulation. The two sharp absorption bands seen for the highly colored and reflective film from the \(\{m = 4, n = 2\}\) formulation indicate the presence of perovskite phases containing one or two layers of Pb–I octahedrons. The weaker, broad, highly red-shifted band at around 750 nm is associated with a quasi-3D \((n \to \infty)\) phase, indicating a degree of disproportionation has occurred in this stoichiometrically n

![Figure 2. Skeletal formulas and abbreviations for the linear-chain alkylammonium salts used in this work.](Image)
formulation. The brown films formed from the \( \{ m = 4, n = 3 \text{ } 5 \} \) formulations show high specular reflectance with minimal light scattering, suggesting uniform compact grains, and their UV–vis absorption spectra indicate the presence of a variety of low-dimensional and high-dimensional (quasi-3D) perovskite phases. The films from the \( \{ m = 4, n > 5 \} \) formulations are nonuniform with a relatively high degree of light scattering and, as expected, progressively come to more closely resemble the 3D MAPbI\(_3\) film with increasing \( n \) value; the gray color of the film from the \( \{ m = 4, n = 8 \} \) formulation and its UV–vis absorption spectrum are consistent with the presence of a considerable fraction of quasi-3D perovskite phases.
The SEM images in Figure 4a demonstrate the significant role of the chain length in the bulky linear-chain alkylammonium cations in determining grain morphology in the drop-cast films. As seen in the top row of images, the \( \{m = 2, n = 4\} \) formulation produces a film comprising large approximately circular grains and the films from \( \{m = 3–5, n = 4\} \) formulations appear to consist of small relatively compact grains, whereas holes and cracks can be seen in the film from the \( \{m = 6, n = 4\} \) formulation. This trend correlates closely with that of the optical characteristics described above, where the films for the \( \{m = 3–5, n = 4\} \) formulations display higher specular reflectance and lower light scattering than the other two \( m \) values. The second row of SEM images in Figure 4a also show that compact perovskite grain morphology is produced for films from the \( \{m = 4, n = 1–5\} \) formulations, whereas large gaps between larger grains are observed for the films of the \( \{m = 4, n = 6–8\} \) formulations. This trend correlates closely with that of the optical characteristics described above, where films from the \( \{m = 4, n = 1–5\} \) formulations display higher specular reflectance and lower light scattering. The needle-like grains seen in the SEM image of the drop-cast 3D MAPbI\(_3\) perovskite film are similar to those reported for spin-cast MAPbI\(_3\) films produced without use of antisolvent or additives. However, the difficulty in the optical characteristics of the film from the \( \{m = 4, n = 8\} \) formulation and the MAPbI\(_3\) perovskite films, the presence of the C\(_4\) alkylammonium cation in the 2DRP perovskite formulation evidently results in a significantly different grain morphology to that in the 3D MAPbI\(_3\) film, suggesting that the bulky alkylammonium cations disrupts the growth of the needle-like MAPbI\(_3\) crystals. The next step of the screening process involves verification that the optical and morphological trends observed in the drop-cast films translate to the photovoltaic (PV) performance of PSCs. Perovskite layers drop-cast from \((C_nA)_2\text{MA}_{m+1}\text{PbI}_{3n+1}\) solutions were incorporated as the light-absorbing layer in devices having the architecture ITO/PEDOT:PSS/perovskite/PC\(_{60}\)BM/PEIE/Ag, where all solution-processed layers were fabricated in air under ambient laboratory conditions (see the Experimental Details section). Considering first the effect of \( m \) on the PV performance, it can be seen from the results in Figure 4b and Table S1 that the devices comprising a drop-cast layer of the \( \{m = 2, n = 4\} \) formulation perform poorly, consistent with its optical and morphological characteristics described above. Devices comprising a perovskite layer based on bulkier alkylammonium cations with \( m = 3–5 \) showed significantly higher PCE, consistent with their more favorable grain morphology. Increasing the size of the bulky cation further to \( m > 5 \) results in loss of PCE and electrical shorting, attributed to the presence of the pinholes seen in the perovskite layer in Figure 4a. This dependence of PV performance on \( m \) correlates very well with the trends in the optical properties and grain morphology of the drop-cast films. It was also found that the rings on the films shown in Figure 3 have a negligible impact on PCE. The effect of the magnitude of \( n \) on PV performance of devices comprising a drop-cast layer of the \((C_nA)_2\text{MA}_{n+1}\text{PbI}_{3n+1}\) formulation can be seen in Figure 4c and Table S2. The poor PV performance of the device produced with \( n = 2 \) is ascribed to the large optical bandgaps of 2DRP phases in these films, as seen in their UV–vis absorption spectra (Figure 3c). Perovskite layers made by using \( n = 3–5 \) formulations give devices showing the highest PCE, consistent with their compact film morphology as shown in Figure 4a. The optimized thicknesses of the perovskite layers are \( \sim 350 \) nm. Significantly lower PCEs were obtained for devices based on formulations having \( n = 6 \) or 8, which is attributed to shorting caused by pinholes in the perovskite layer. Little hysteresis was observed in the \( J–V \) curves measured in forward and reverse scans (Figure S1), and the steady-state PCE of the “champion” cell \((\{m = 4, n = 5\})\) formulation recorded by using maximum power point (MPP) tracking (Figure S2) of 15.04% is consistent with the \( J–V \) measurement. As expected from the film morphology of the drop-cast MAPbI\(_3\) film, PSCs comprising a drop-cast layer of MAPbI\(_3\) showed extremely poor PV performance.
formulations are shown in Figure 6 as a function of \( n \). Like the UV−vis absorption spectra of the drop-cast films, the PL spectra of these films also display bands signaling the presence of a variety of 2DRP perovskite phases (Figure 5). The film produced by using the \( n = 1 \) formulation displays a single, strong, sharp PL band irrespective of which side is probed, confirming that this film comprises solely 2DRP perovskite phase comprising a single Pb−I octahedron layer. While various mechanisms have been proposed to describe the phase distribution in films produced by using 2DRP perovskite formulations,\(^{36−38}\) the general consensus is that the 3D or quasi-3D perovskite phases initially form at the air−liquid interface during the drying process, with the bulky alkylammonium cations forced toward the substrate−liquid interface where there is preferential formation of lower-dimensional perovskite phases.\(^{37}\) However, the complete absence of the broad, red-shifted PL band at \( \geq 750 \) nm associated with quasi-3D perovskite phases for films produced from (C\(_4\)A\(_2\))\(_n\)−1Pb\(_n\)I\(_3\)\(_n\)+1 formulations having \( n < 4 \) is intriguing (Figure 6).

Evaporation of solvent initially causes a higher solute concentration at the air−liquid interface than in the bulk solution, as illustrated in Figure 5b. Consequently, crystal nucleation and growth tend to begin at the air−liquid surface.

Figure 5. Illustrations of crystal growth in the solution of MAPbI\(_3\) perovskite and 2DRP perovskites containing bulky cations (a), the solute concentration gradient in the solution caused by solvent evaporation (b), and large cations (e.g., C\(_m\)A) in the solution of 2DRP perovskite formulations (c).

Figure 6. PL emission spectra of films drop-cast from solutions of (C\(_4\)A\(_2\))\(_n\)−1Pb\(_n\)I\(_3\)\(_n\)+1 in DMF, as a function of \( n \). Peak numbering corresponds to the number of Pb−I octahedron layers in the various 2DRP phases in the films. Samples consisted of glass/PEDOT:PSS/perovskite. Excitation and emission detection by using the perovskite (front) side or the glass (back) side, as indicated (excitation wavelength: 440 nm).
A critical MA:Ca molar ratio in the 2DRP perovskite formulation appears to exist, below which the formation of low-dimensional perovskite phases dominates. In the specific case of CaA cations, the critical MA:Ca ratio is found to be 2, with higher-dimensional quasi-3D phases observed only when \( n \geq 4 \). For example, for \((C_{4}A)_{2}MA_{n}Pb_{5}I_{16}\) formulations with \( n \geq 4 \), the initial concentration of MA in the drop-cast solution is at least twice that of CaA, and in this case the quasi-3D perovskite phases appear to have a competitive advantage for formation at the air–liquid interface. The resulting higher depletion rate of MA than CaA at this interface then leads to a concomitant decrease in the MA:Ca ratio in the bulk solution, resulting in enhanced formation of low-dimensional phases once the MA:Ca ratio falls below 2. A prominent red-shifted PL emission band centered around 760 nm, associated with the quasi-3D phases, is seen in the spectra of films from the formulations with \( n \geq 4 \) irrespective of which side of the sample is probed, indicating that the quasi-3D phase perovskites are distributed throughout the drop-cast film. In contrast, the absence of PL emission from low-dimensional perovskite phases upon excitation and detection from the perovskite side of the sample confirms that these phases are concentrated more closely to the glass–perovskite interface. Notably, when \( n = 8 \), the excess concentration of MA over CaA throughout the film forming process appears to be sufficiently large to completely suppress the formation of low-dimensional phases.

The spectral distribution of PL emission from drop-cast films of \((C_{n}A)_{2}MA_{n}Pb_{5}I_{16}\) \((n = 4)\) formulations was found to be essentially independent of the size of the CaA cation, with the exception of the \( n = 2 \) (ethylammonium) cation (Figure S5). The absence of PL bands and UV–vis absorption bands associated with low-dimensional phases for this film indicates that it comprises only quasi-3D perovskite phases, suggesting that the CaA cation is not sufficiently bulky to induce the formation of a low-dimensional perovskite phase. Unlike the other perovskite films, perovskite film prepared by using CaA shows Ruddlesden–Popper perovskite structures \((n = 2, n = 3)\) both at the surface and bottom of the film (Figure S5f). This may be attributed to the long hydrophobic carbon chain which is capable of dwelling at the surface of the solution acting similar to the surface surfactant (Figure 5c), leading to the increased concentration of CaA at the surface and in turn inducing the formation of Ruddlesden–Popper perovskites.

A comparison of the trends in device PV performance in Tables S1 and S2 and the perovskite phase information obtained from PL and UV–vis absorption spectroscopy suggests that the coexistence of a mixture of lower- and higher-dimensional (quasi-3D) perovskite phases in the drop-cast films may be important for good device performance. Apart from the benefits to grain morphology described above, charge transport in low-dimensional perovskite phases alone is known to be highly dependent on crystal orientation,36 and so it may be that their coexistence with quasi-3D perovskite phases boosts the efficiency of transportation of charge carriers to the electrodes, leading to improved device performance.

The results of the screening method described above indicate that the drop-cast film displaying the best overall characteristics was produced by using the \((C_{4}A)_{2}MA_{4}Pb_{5}I_{16}\) formulation \((m = 4, n = 5)\). The PV performance of this optimal formulation was, therefore, evaluated as a slot-die coating on a flexible substrate by using a roll-to-roll (R2R) process (Figure 7a). Two hot plates were used to heat the film, as was the case for drop-casting. A first hot plate at 60 °C was used to dry the film (see Figure S6), followed by a second hot plate at 100 °C for thermal annealing. A photo of the R2R slot-die-coated perovskite film on the flexible PET/ITO substrate is shown in the inset of Figure 7b. As found for the drop-cast film, the R2R slot-die-coated film shows a uniformly high specular reflectance and is pinhole-free, with a thickness of \( \sim 350 \) nm. Perovskite solar cells were prepared by using this slot-die-coated perovskite film, yielding a “champion” cell with \( V_{oc} = 1.08 \) V, \( J_{sc} = -15.58 \) mA/cm\(^2\), \( FF = 52.02\%\), and PCE = 8.75% (Figure 7b). Although this PCE is comparable with other values reported for devices based on R2R-coated perovskites,29 it is substantially lower than that of the cells having the same active area prepared by using a drop-cast layer of the same 2DRP perovskite formulation on glass/ITO substrates (Tables S2 and S3). This is attributed to the substantially higher sheet resistance and roughness of the flexible PET/ITO substrate than the glass/ITO substrates.

**CONCLUSIONS**

In summary, simple drop-cast film preparation under ambient laboratory conditions, coupled with visual inspection and routine optical, microscopy, and electrical measurements, was shown to provide a facile method for effectively screening an array of 2D organic–inorganic hybrid lead–halide Ruddlesden–Popper perovskite formulations for their potential to form high quality films for use in solar cells. A mechanism was proposed to explain the dependence of perovskite grain morphology and distribution of the multidimensional perovskite phases on the 2DRP perovskite formulation used for drop-casting.
Screening of drop-cast films enabled the number of compositions of potential interest to be substantially reduced. Ultimately, the optimally performing formulation was verified by fabricating and testing devices, but screening meant that the number of devices needing to be evaluated in this final step was greatly reduced. The similarity between the drying mechanisms for drop-cast and slot-die coated films, and the greater simplicity of the drop-cast method, means that this screening offers a quick and useful guide to likely quality of a slot-die-coated film for a given 2DRP perovskite formulation. The drop-casting method has the merit of simpleness, with the demerit of scalability. The compatibility of the drop-casting method has the merit of simplicity of the drop-cast method, means that this screening enabled the number of devices needing to be evaluated in this final step was greatly reduced. The higher sheet resistance and surface roughness of the underlying flexible PET/ITO substrate are likely to be responsible for the PCE being substantially lower than that for devices comprising a drop-cast layer of the same perovskite formulation on a glass/ITO substrate.

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