

An exploratory data analysis on certified perovskite devices efficiency and I-V metrics: insights into materials engineering and process scaling up

Jialiu Ma* and Da Guo

Advanced research, First Solar, Inc. Perrysburg, Ohio 43551, USA

Abstract: Perovskite based solar cells have achieved tremendous progress reaching record efficiency in the past 5 years. Numerous new processes and chemistry have been reported and contribute to the perovskite rapid progress. Continuous efficiency improvements are still necessary for perovskite solar cells, and an exploratory data analysis on devices performance over multiple studies could boost the technology development. Such analysis could identify patterns or provide insights that are not obvious in a single study. Here we present a high quality dataset containing only independently certified Pb-based perovskite solar cells summarizing their efficiency, relevant I-V metrics, manufacturing processes and materials used. Analysis over the dataset provides insights on how aperture size, perovskite deposition methods and materials used in each functional layer affect the final solar cell efficiency and I-V metrics. Future directions are also suggested for efficiency improvements.

Keywords: Perovskite, Solar cell, scale up, efficiency, certification, process.

Perovskite solar cells have seen tremendous improvements in the past several years¹. Their efficiency have matched or even exceeded the state of art inorganic photovoltaic devices that have gone through decades of research and development². Continuous efficiency improvements are crucial for perovskite solar cells. Higher efficiency will enhance the cost advantage towards commercialization³. Thus the quest for new record of device efficiency and relevant I-V metrics (open-circuit voltage (Voc), short-circuit current density (Jsc) and Fill Factor (FF), etc.) continues in the perovskite community. Despite the large number of perovskite solar cell papers published since 2013, advancing towards a higher efficiency perovskite device could be a daunting task for the following reasons.

- Process development sometimes relies solely on empirical knowledge from a small number of studies with limited processes/materials tested⁴. Such empirical knowledge can be misleading regarding what process/materials should be focused on for further optimization.
- Device data quality in the literature differs from lab to lab. Lateral comparison across different studies are challenging due to lacking widely accepted standards for measuring and reporting device performances⁵⁻⁷. While important lessons can and are learned within the perovskite community, finding common themes across studies is critical to determining if observations are general and cross cutting or narrow and specific.
- The majority of research lab level efforts are still focused on cell level that lacks a connection to process scaling up⁵. This poses obstacles for the commercialization of perovskite solar cell technology.

Exploratory data analysis is an approach that combines the results of multiple scientific studies. This tool could identify patterns or provides insights that are not obvious in a single study. Furthermore, the collection of data across multiples studies on the same process/material could lead to a more statistically solid conclusion. Hereby, we present an exploratory data analysis on a high-quality perovskite efficiency dataset containing only independently certified Pb-based perovskite solar cell devices. The analysis does not intend to provide a comprehensive review over efforts on improving perovskite solar cell efficiency

which can be found in other excellent reviews^{1,8-12}. Instead, this analysis focuses on revealing trends and insights that is obscured in single study regarding process scaling up, device fabrication methods, components engineering and their impact to efficiency and I-V metrics. The analysis would provide the following to address challenges towards new record perovskite devices:

- A high-quality perovskite solar cell data set that only contains independently certified devices. Information on efficiency, I-V metrics, materials and processes used for fabricating the devices are summarized for each individual study.
- An exploratory data analysis focusing on the following aspect:
 1. Temporal evolution of perovskite solar cells device efficiency and I-V metrics
 2. Effects of scaling up device aperture size on efficiency and I-V metrics
 3. Effects of spin coating vs non-spin coating perovskite deposition method on efficiency and I-V metrics
 4. Effects of types of transparent conductive oxide (TCO) substrates used (Fluorine doped tin oxide (FTO) vs Indium doped tin oxide (ITO)) on efficiency and I-V metrics
 5. Effects of perovskite absorber chemistry on device efficiency and I-V metrics
 6. Effects of types of electron transport layer (ETL)/hole transport layer (HTL) used on efficiency and I-V metrics
- Suggestions based on the exploratory data analysis for future development directions on materials selection, device processing, scaling up and fundamental understanding.

It is a known challenge to measure perovskite device performances consistently, accurately and precisely due to issues such as device hysteresis and environmental instability¹³. Analysis on poor measurements data lacking environmental control (temperature and humidity), well-maintained electrical setup and well-calibrated light source can lead to biased conclusions. In this paper, only device data that is verified by independent certification agencies are included for analysis. The certification agencies are meticulous about all aspects of measurement (setup, tools, light source, electronics, environmental control, etc.) and comply with international standardization requirements. Furthermore, certification reports containing original data are available in the publications making data management feasible.

To the best of our ability, we collected 69 certified devices in a total of 63 publications in the public domain from 2013 to 2020¹⁴⁻⁷⁶. Certification report in each publication is digitized and efficiency, I-V metrics (Voc, Jsc and FF) and measurements settings are summarized into a single table. We also extract device fabrication and materials information from those publications and add in the table. The table is formatted as an accessible CSV format for the perovskite community to use and analyze. The number of publications reporting certified devices has increased steadily from 1 publication in 2013 to 18 in 2019. These devices are manufactured from a wide geographic origins of 12 different countries and 38 institutions. Among the certification agencies, Newport PV lab has certified most number of devices (34 out of 69 devices).

Scaling up the size of the perovskite solar cell is a necessary step towards future commercialization. Larger solar cells or modules usually demonstrate lower efficiency due to scaling up issues³. Thus it is crucial to look at the device size when comparing devices manufactured from different chemistry and processes. Furthermore, the defined area of a solar cell during measurements has a large effect on the efficiency. Processes such as laser scribing and bussing will add in non-functional dead areas for solar modules. The certified devices typically report the aperture area which is defined by masking and include all components such as active material, busbars, fingers and interconnects. We will use aperture area in this paper to represent the total cell or module size. All certified devices are divided into three groups based on aperture area. Among the 69 certified devices, 47 devices' aperture area are close to 0.1 cm²

(termed sub-cell device in this paper) and 14 other devices close to 1 cm^2 (termed cell device in this paper). Aperture area $<0.1 \text{ cm}^2$ are still the most adopted aperture area for new perovskite technology demonstration. Only 8 certified devices were reported with an aperture area over 10 cm^2 (termed submodule device in this paper). Included in our dataset, the largest certified device was reported with an aperture area of 63.7 cm^2 , close to an $8 \text{ cm} \times 8 \text{ cm}$ size. New perovskite technology breakthrough demonstrated and certified at a submodule level are still relatively rare.

Perovskite solar cell efficiency improved rapidly in the past 5 years¹. A temporal analysis of the efficiency data can provide the improvements trend. Certified perovskite solar cells reported before 2016 mostly show efficiency below 20%. Only after 2016, sub-cell devices consistently exceed 20% efficiency (Fig.1). The 47 sub-cell devices are certified with efficiency ranging from 11.01% to 23.48% with a median efficiency of 20.50% (Fig.2a). Scaling up device size does decrease their efficiency. It took another 2 years until 2018 before the first certified cell device to reach 20% efficiency mark. Among the 14 certified cell devices, the lowest efficiency is 6.59% and highest being 20.87% with a median of 19.30% (Fig.2a). Scaling up from 0.1 cm^2 aperture area to 1 cm^2 lead to an absolute 1.18% median efficiency drop. Attempts to further scale up the solar cell size show greater efficiency drop. The first certified submodule size device was reported in late 2016 with an efficiency of only 9.11%. The efficiency improved substantially in the past 3 years with the newest submodule device certified with an impressive 16.50% efficiency (Note: NREL efficiency chart has recently revealed a submodule device certified with 17.25% efficiency from Microquanta, Inc². However, the details of the device fabrication and processing are not available in the public domain, so it is not included in our analysis). The median efficiency of the 8 submodule devices is 14.69%, 5.81% lower than the sub-cell size devices median efficiency.

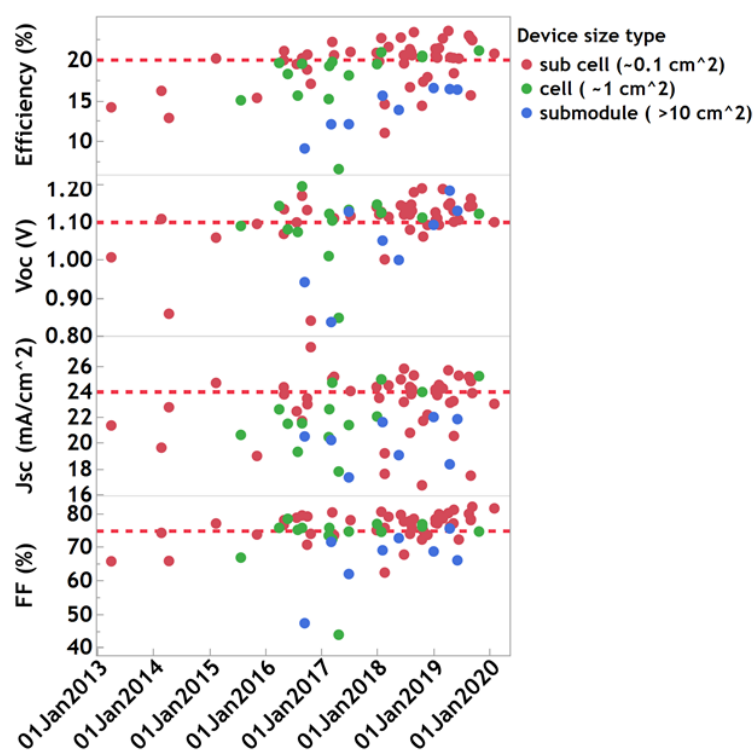


Fig1. Certified perovskite's solar cell efficiency, Voc, Jsc and FF plotted by publication submission date and colored by their aperture areas (Red: sub cell device; Green: cell device; Blue: submodule device).

Red dotted lines marking 20% Eff, 1.10V Voc, 24 mA/cm² Jsc and 75% FF are drawn for reference purpose.

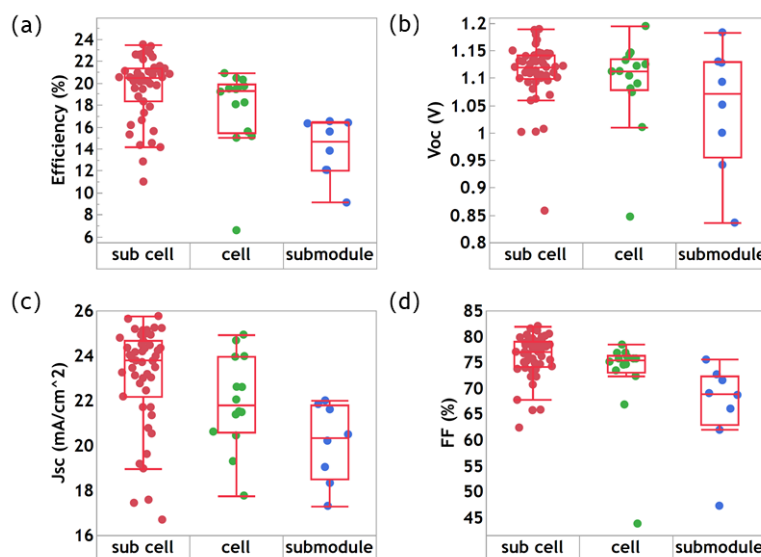


Fig2. (a) Efficiency, (b) Voc, (C) Jsc and (d) FF from all certified perovskite solar cells with different aperture areas (sub cell: ~0.1 cm², cell:~1 cm², mini-module: >10 cm²). Upper and lower boundaries of the boxes represent the third quartile and first quartile, respectively. Red error bars indicate 1.5 times interquartile range out from third quartile and first quartile, respectively. The red middle line in each box indicated the median value.

Basic parameters determined from I-V curves under standard test conditions such as Voc, Jsc and FF provide more details on the device improvement. An analysis of these parameters could help understand the efficiency gap for device process scaling up¹³. Both sub cell and cell size solar cells show a median Voc over 1.1 V with a narrow distribution (median Voc: sub-cell devices:1.12 V; cell devices:1.11V). Submodule solar cells have a much wider distribution of Voc with only 3 out of the 8 devices' Voc exceeding 1.1V (Fig 2b). On the other hand, Jsc for scaled up devices are substantially lower (median Jsc: sub-cell device: 23.77 mA/cm²; cell devices: 21.77 mA/cm²; submodule devices: 20.34 mA/cm²) (Fig. 2c). In addition to Jsc differences, a small fill factor gap is found even for most recent devices (median fill factor: 77.0% for sub-cell device vs 75.4% for cell device vs 68.8% for submodule devices) (Fig.2d). Efforts developing scaling up process for perovskite solar cells should focus on root cause investigations and process/materials engineering to minimize the gap for Jsc and FF.

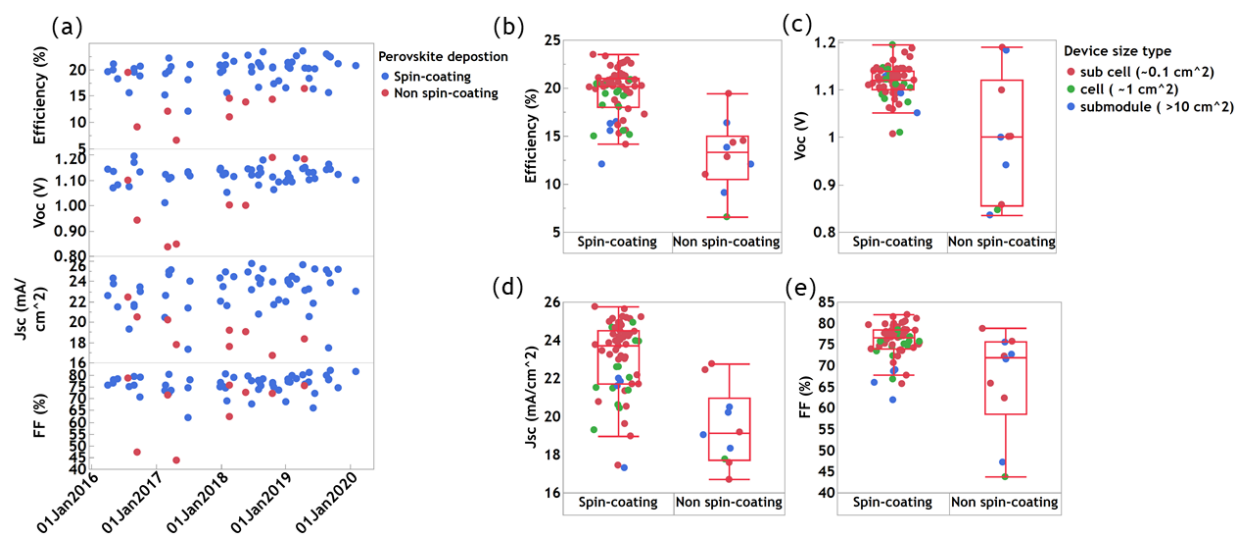
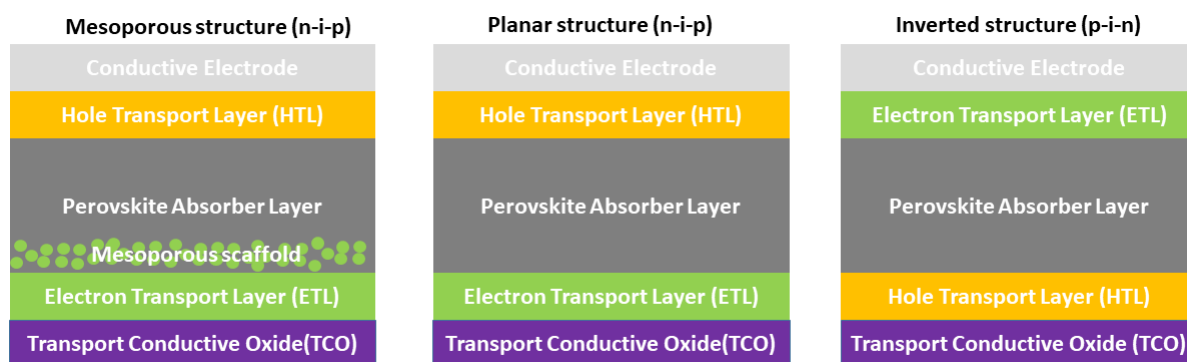


Fig3. (a) Certified perovskite's solar cell efficiency, Voc, Jsc and FF plotted by publication submission date and colored by perovskite deposition method (Blue: spin-coating; Red: Non spin-coating). The (b) Efficiency, (c) Voc, (d) Jsc and (e) FF data are also grouped by perovskite deposition method for comparison. Each device data point is colored by its aperture area size (red: sub cell $\sim 0.1 \text{ cm}^2$, green: cell $\sim 1 \text{ cm}^2$, blue: submodule $> 10 \text{ cm}^2$). Upper and lower boundaries of the boxes represent the third quartile and first quartile, respectively. Red error bars indicate 1.5 times interquartile range out from third quartile and first quartile, respectively. The red middle line in each box indicated the median value.

One critical process to scale up for perovskite solar cell is deposition for perovskite as the absorber layer⁷⁷. Currently spin coating is the most adopted method to deposit perovskite layer in a research lab setting. 60 out of 69 certified solar cells used the spin coating method for perovskite deposition. However, applying spin coating method for high volume manufacturing will be a great challenge. It is needed to experiment with alternative high volume manufacturing friendly methods to deposit perovskite. 10 certified solar cells attempted using non-spin coating methods including screen printing, spray coating, vacuum deposition, pressure processing, blade coating, infiltration and thermal evaporation. It is found that non-spin coating method produce overall lower efficiency solar cells (Fig. 3a, b) (median efficiency: spin-coating 20.17% vs non spin-coating 13.33%) with lower Voc (Fig.3c), Jsc (Fig.3d) and FF (Fig.3e). However, it is unfair to conclude these alternative methods are inferior to spin coating method. Spin-coating method has been intensively developed and optimized for the state-of-art perovskite device. Non-spin coating depositions simply need more method development and process optimization. It is also noticed that hole transport layer and electron transport layer also heavily rely on spin coating method for deposition in almost all certified solar cells. More efforts in searching, screening and optimizing alternative high volume manufacturing friendly methods are needed for both absorber and transport layer depositions processes.



Scheme 1. Three types of perovskite device structure used in certified perovskite solar cells.

A high efficiency perovskite solar cell typically consists of multiple functional layers including transparent conductive oxide (TCO) layer, electron transport layer (ETL), perovskite absorber layer, hole transport layer (HTL) and conductive electrode layer (Scheme 1). The improvements on these components and how they impact on device efficiency have been reviewed comprehensively elsewhere^{1,8-12}. In this paper, we focus on understanding how choice of materials for each layer could impact the efficiency and I-V metrics. As noted before, devices with larger aperture size (cell and submodule size) and those using non spin-coating method for perovskite deposition demonstrate lower efficiency and need more process development and optimizations. So the following analysis for each functional layer will only include sub cell size devices using spin coating method for absorber deposition.

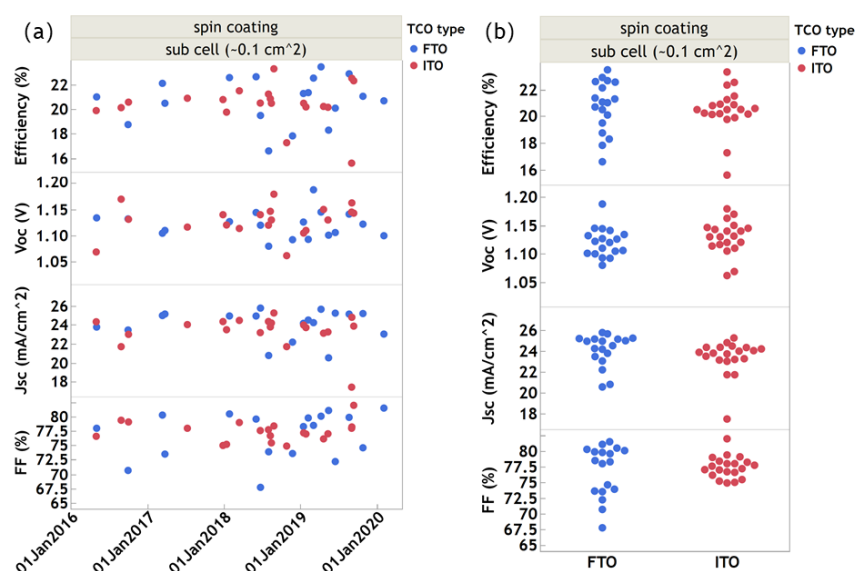


Fig.4 (a) we include only sub cell size solar cell using spin coating to deposit perovskite for analysis. Efficiency, Voc, Jsc and FF data are plotted by publication submission date and colored by TCO used in the device. These aggregated data on efficiency, Voc, Jsc and FF data are shown in Fig.4 (b) for comparison.

Transparent conductive oxide (TCO) layer provides a conductive pathway for electron collection in perovskite solar cells⁷⁸. TCO layer thus needs high transparency and low resistivity. Two types of TCO are used in the certified devices and are both commercially available: fluorine doped tin oxide (FTO) (42 certified devices) and indium doped tin oxide (ITO) (27 certified devices). ITO gained its popularity since

2016 and are more frequently adopted in a planar p-i-n type structure. Temporal data analysis shows that devices adopting either ITO or FTO as the TCO layer could achieve record efficiency (Fig. 4a). Previous studies on TCO used in solar cells show that differences on TCO transparency and resistivity can affect Jsc and FF in theory⁷⁹. However, analysis show small differences in Jsc and FF between devices using different types of TCO (median Jsc 23.81 mA/cm² ITO vs 24.71 mA/cm² FTO; median FF 77.4% ITO vs 78.4% FTO) (Fig.4b). It is noticed that none of the certified device publications were devoted to the TCO layer development and optimization. Future studies on TCO optimization of their properties (transparency, resistivity, band structure alignment and interface) could potentially boost efficiency for new record device.

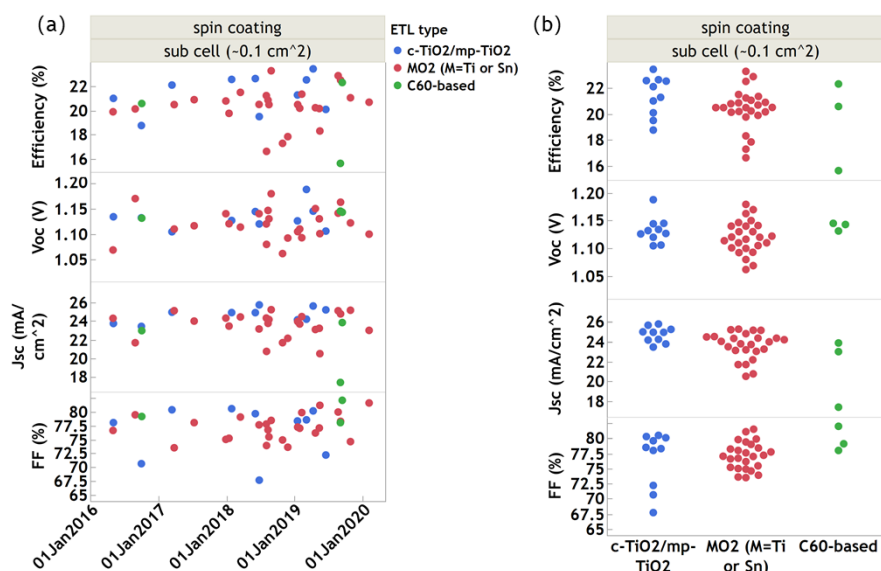


Fig.5 (a) only sub cell size solar cells using spin coating to deposit perovskite are included for analysis. Efficiency, Voc, Jsc and FF data are plotted by publication submission date and colored by ETL used in the device. The grouped data are shown in Fig.5 (b) for comparison.

Electron transport layer functions in extracting photo-generated electrons and aligning interfacial energy level⁸⁰. Mesoporous TiO₂ was exclusively used as ETL in certified perovskite solar cells before 2016. Afterwards, non-porous SnO₂ and TiO₂ gradually gained popularity. Among the 69 certified devices, 28 devices use a combination of a compact (non-porous) TiO₂ layer and a mesoporous TiO₂ layer as scaffold for perovskite absorber. 31 devices only use nonporous metal oxide such as SnO₂ or TiO₂ as the electron transport layer. Alternative organic based materials such as C₆₀ derivatives are also demonstrated in the rest 11 solar cells (Fig.5a). Analysis shows that the non-porous metal oxide based ETL solar cells efficiency is lower to those using mesoporous ETL layers (median Eff: non-porous ETL devices 20.5% vs mp-TiO₂ ETL devices 21.7%). However, the record efficiency in these two types ETL produce devices are closely matched (maximum efficiency: non-porous ETL devices 23.32% vs mp-TiO₂ ETL devices 23.48%) (Fig. 5b). Both type of ETL could potentially achieve the record efficiency.

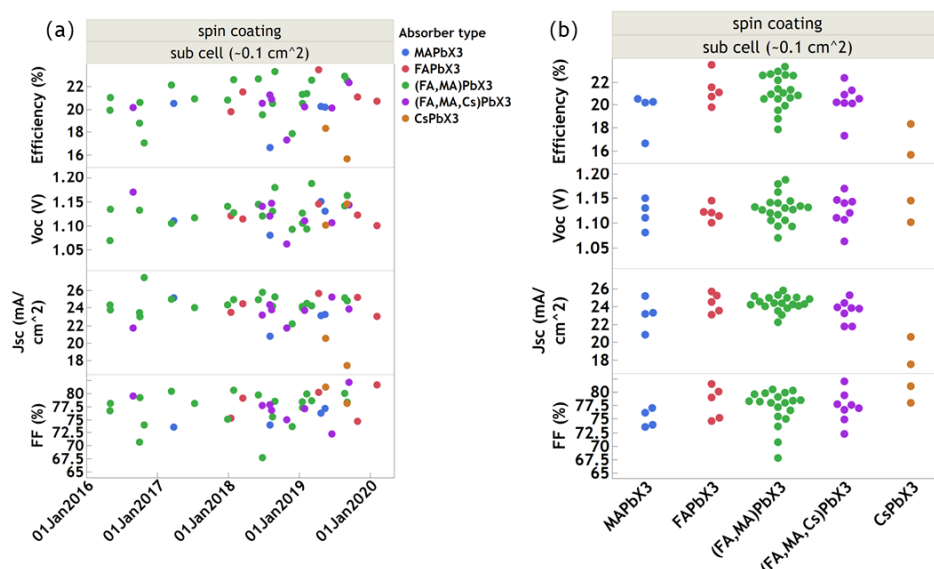


Fig.6 (a) only sub cell size solar cells using spin coating to deposit perovskite are included for analysis. Efficiency, Voc, Jsc and FF data are plotted by publication submission date and colored by absorber chemistry used in the device. The grouped data are shown in Fig.6 (b) for comparison.

The perovskite absorber layer absorb photons and converted them to carriers for extraction¹. It is the single most critical component in a perovskite solar cell. In the very first perovskite solar cell report, CH₃NH₃PbBr₃ (MAPbBr₃) and CH₃NH₃PbI₃ (MAPbI₃) were investigated as a sensitizer for solar cells⁸¹. Following that report, anion (Br⁻ in the case of MAPbBr₃) and cation (MA⁺ in the case of MAPbBr₃) chemistry are heavily investigated for perovskite composition⁸². These researches contribute to the rapid perovskite solar cell efficiency improvements. It is noticed that the practice of mixed bromine and iodine anion composition is adopted among nearly all certified solar cells. This practice enables band gap tuning to optimize the light absorbance over the full visible light spectrum. Turning to cation composition, it is observed that cation MA could be partially substituted by FA or a combination of FA and inorganic cation such as Cs⁺, Rb⁺ or K⁺. The above anion and cation compositional engineering combined lead to five major compositions among the certified perovskite solar cells: MAPbX₃ (X=I,Br) (20 devices), FAPbX₃ (X=I,Br) (5 devices), (FA,MA)PbX₃ (X=I,Br) (29 devices) and (FA,MA,Cs)PbX₃ (X=I,Br) (13 devices) and CsPbX₃(X=I,Br) (2 devices). Out of all 69 certified solar cells, more than 90% use the following three formulations: MAPbX₃ (X=I,Br), (FA,MA)PbX₃ (x=I,Br) and (FA,MA,Cs)PbX₃ (X=I,Br). An comparison over the three most used formulation show that partially substituting MA with FA improves the overall efficiency (median efficiency MAPbX₃ vs (FA,MA)PbX₃: 20.20% vs 21.02%) (Fig. 6a). The efficiency improvement is mostly gained through Jsc (median Jsc MAPbX₃ vs (FA,MA)PbX₃: 23.17 mA/cm² vs 24.31mA/cm²) and FF (median FF MAPbX₃ vs (FA,MA)PbX₃: 75.0% vs 78.2%). However, adding Cs to (FA,MA)PbX₃(X=Br,I) does not further improve the efficiency (median Eff (FA,MA)PbX₃ vs (FA,MA,Cs)PbX₃ 21.0% vs 21.0%) (Fig. 6b). Recent literature indicated the role of Cs are more likely related to the overall stability improvement of perovskite based device⁸³. The stable and active perovskite phase of FAPbX₃ (α phase) and CsPbX₃ (β phase) has previously been a challenge to synthesize and prone to phase degradation at room temperature due to the large cation size^{58,71}. Only until very recently these phases are successfully obtained and certified. It is worth noting that FAPbX₃ based devices could reach efficiency comparable to the devices based on (FA,MA)PbX₃ with matching Voc, Jsc and FF.

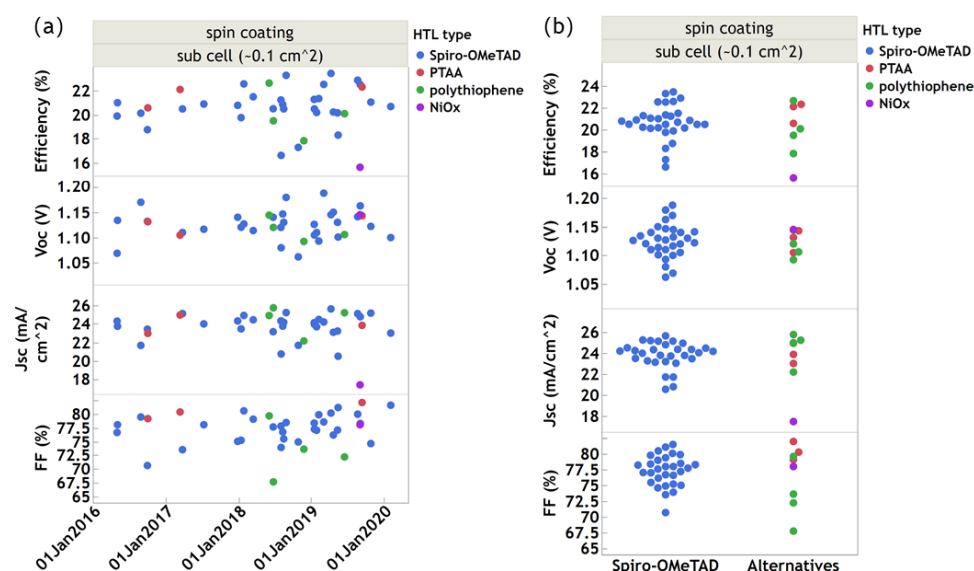


Fig.7 (a) only sub cell size solar cells using spin coating to deposit perovskite are included for analysis. Efficiency, Voc, Jsc and FF data are plotted by publication submission date and colored by HTL used in the device. The grouped data are shown in Fig.7 (b) for comparison.

Hole transport layer transports the photogenerated holes towards conductive electrode and align the interface band energy with the perovskite absorber⁸⁴. One single material, 2,2',7,7'-Tetrakis[N,N-di(4-methoxyphenyl)amino]-9,9'-spirobifluorene (spiro-OMeTAD), is prevalently used as HTL in perovskite solar cells. 44 out of 69 devices used spiro-MeOAD as the hole transport layer with the addition of lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) and 4-tert-butylpyridine (tBP) to improve its conductivity. A few other types of materials such as polythiophene, poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine] (PTAA) and NiO_x are also being explored. An analysis of the I-V metrics HTL materials used shows that alternative HTL material can achieve a similar efficiency compared to the commonly used spiro-OMeTAD (Fig.7a,b). PTAA and polythiophene-based HTL could result solar cell efficiencies similar to those based on spiro-OMeTAD. Recent literature suggested that spiro-OMeTAD could lead to lower device stability due to moisture ingress⁸⁵. Efforts are needed for exploring and optimizing alternative HTL materials without compromising the overall device efficiency and stability.

The analysis on the device aperture size, perovskite deposition method and the components materials reveals insights that are not obvious from single perovskite study. There are still great challenges ahead for scaling up the size of perovskite device and achieve high efficiency for commercial success. Deposition method is only one of the obvious issues in device process scaling up. Alternative methods to spin coating for the deposition of perovskite and HTL/ETL layers needs to be systematically explored and optimized before the final assessment for the best approach. Other issues such as device uniformity, laser scribing and encapsulation all need to be considered for scaling up. Equally important is the assessment on materials and chemistry for device structure and all functional layers. Many other alternatives could potentially be more beneficial on efficiency and other performance metrics (stability, temperature coefficient, etc.). The widespread use of a certain material could simply be traced back to historical reason. A more systematic experimental assessment on all previously demonstrated materials in perovskite device could be planned in the near future.

Supporting Information

The certified perovskite solar cell data set is available online as a single file.

Corresponding Author

Email: majiali@gmail.com

Notes

The authors declare no competing financial interest

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